Time-averaged and time-resolved laser optical temperature measurements in water with Filtered Brillouin Scattering combined with LDV

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ABSTRACT

A laser measuring system was developed and built that allows to optically measure temperature in water using the method of Filtered Brillouin Scattering (FBS). First time-resolved optical temperature measurements were demonstrated. Furthermore, the FBS-system was combined with an LDV to enable simultaneous measurement of flow velocity and therefore the system is also capable to measure the heat flow.

Time-averaged temperature values were determined with good accuracy and, as a special highlight, also time-resolved temperature measurements have been demonstrated with temporal resolution in the order of approximately 10 ms, validated by comparison with fast thermocouple measurements.

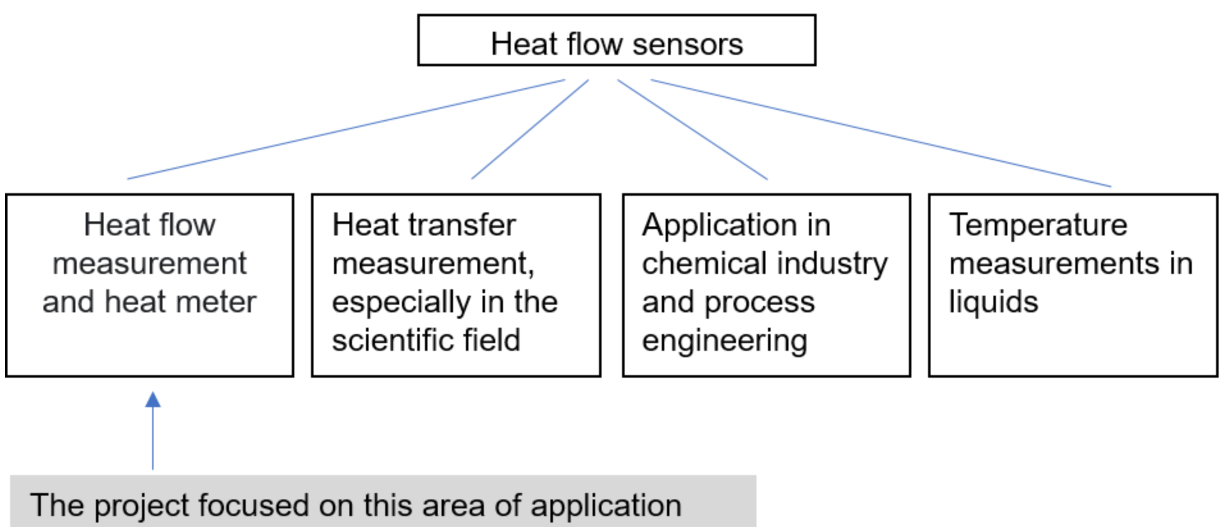
The overarching goal of the research project was to prepare the market introduction of a laser-optical measuring system for spatially point-based and time-resolved measurement of the heat flow in liquids, especially in water. In order to realize this, it was necessary to measure the local velocity and the local temperature in a liquid. The optical measurement of the local velocity has long been possible using the established method of laser Doppler velocimetry (LDV). Therefore, the heat flow measurement method to be developed should be based on this technology. Until now, there was no suitable optical method for measuring the temperature. In recent years, however, the physical phenomenon of Brillouin scattering has become one focus of measurement technology development. If a small volume of liquid is irradiated with light, the molecules in the liquid scatter back part of the light, which is known as Brillouin scattering. The spectrum of the scattered light depends on the local temperature in the liquid; and it turns out that this physical relationship can be exploited to develop a highly accurate, fast, and non-contact method for measuring temperature.

In this paper, we explain the Filtered Brillouin Scattering (FBS) method, show a setup for measuring temperature and velocity in water flows and thus a method for determining the heat flow, and demonstrate the measurement accuracy using a calibration test bench. The temperature measurement accuracy achieved is in the order of 1 K.

1. Introduction

Heat flows in moving liquids play a crucial role in many technical areas, such as process and process engineering, the chemical industry, heating applications both domestic and industrial, in the investigation and optimization of heat transfer processes, in heat flow measurement and in the operation of a heat meter[1]. However, the heat flows in such processes can only be measured imprecisely because there is a lack of high-precision sensors. With today's measurement techniques, one is only able to determine heat flow as an average over time, with averaging over large spatial volumes as well. This method is inaccurate. It is therefore necessary to develop a sensor technology with which the heat flow can be measured with high spatial and temporal resolution. At the same time, this measurement technology should work without contact in order not to falsify the measurement result.

In order to measure the heat flow in a liquid at a specific point, both the local velocity and the temperature must be recorded. The fields of application of the heat flow sensor can be roughly divided as follows:



Heat flow measurements in large pipe diameters are particularly necessary in district heating networks. So far, the volume flow averaged over time has been measured, as well as the temperatures in the upstream and downstream section at single locations. The temperature sensors used usually have a response time in the order of seconds. The heat flow is calculated from the mass flow (or the volumetric flow together with the liquid's density), the temperature difference between upstream and downstream, and the heat capacity of the fluid. Correlations

[1] A heat meter is a device that determines consumption by measuring the heat flow in the flow and return of a consumer .

between temperature and speed fluctuations cannot be recorded, which can lead to significant errors.

The experience of Optolution GmbH from the on-site calibrations of large flow sensors (DFS) in district heating systems show that more than a quarter of the calibrated DFS showed measurement deviations under operating conditions that were greater than the so-called traffic error. Depending on the class (essentially determined by the measuring principle used) of the measuring devices, the traffic error limits are +/- 4% (class 2) or even +/- 6% and are therefore of a considerable magnitude in themselves.

Since very large amounts of heat are transferred through district heating networks, but measurements so far can demonstrably have large errors, increased measurement accuracy can provide a fairer billing of the transferred heat.

The novel heat flow sensor presented here combines the established optical speed measurement of laser Doppler velocimetry (LDV) with an innovative temperature measurement technology based on so-called Brillouin scattering. Retrofitting existing LDV devices is possible.

2. Optical access to the measuring point in district heating pipes

At least one optical access point to the pipeline with the flowing medium is required for the measurements. For the application of the measuring method in practice, there is an access construction that can be installed without interrupting the operation of the system and allows the inner window to be replaced at any time if it is soiled. This construction, including the process and the device for installing and removing the windows under operating conditions, is patented (inventors and owners: ILA R&D GmbH and Optolution Messtechnik GmbH). Such optical accesses to pipelines have been installed in the field approximately a hundred times as part of on-site calibrations of large flow sensors in district heating transfer stations, district heating generation plants and drinking water systems. Figure 1 through Figure 2 illustrate the solution.

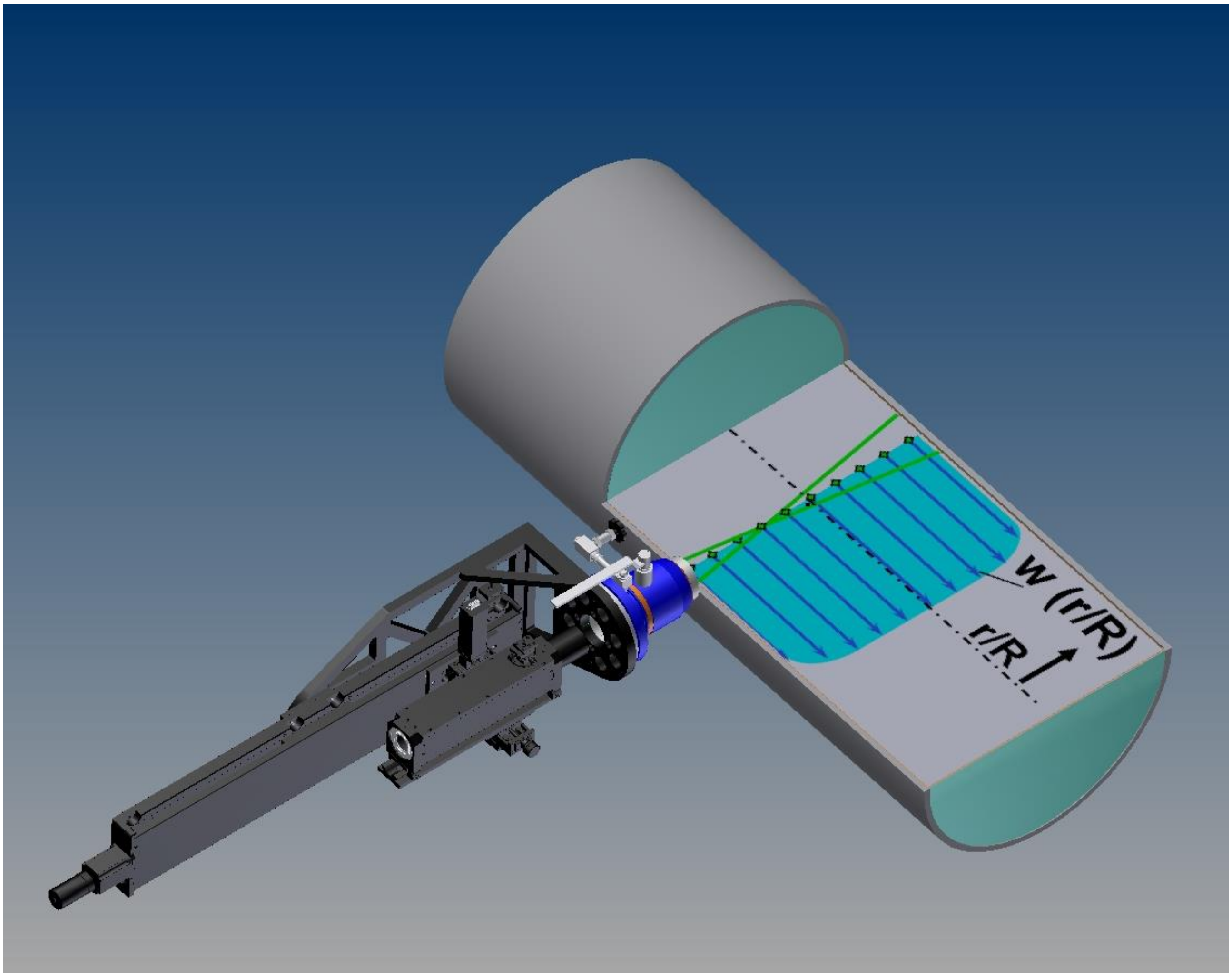


Figure 1. Pipe with water flow and LDV on traversing unit.

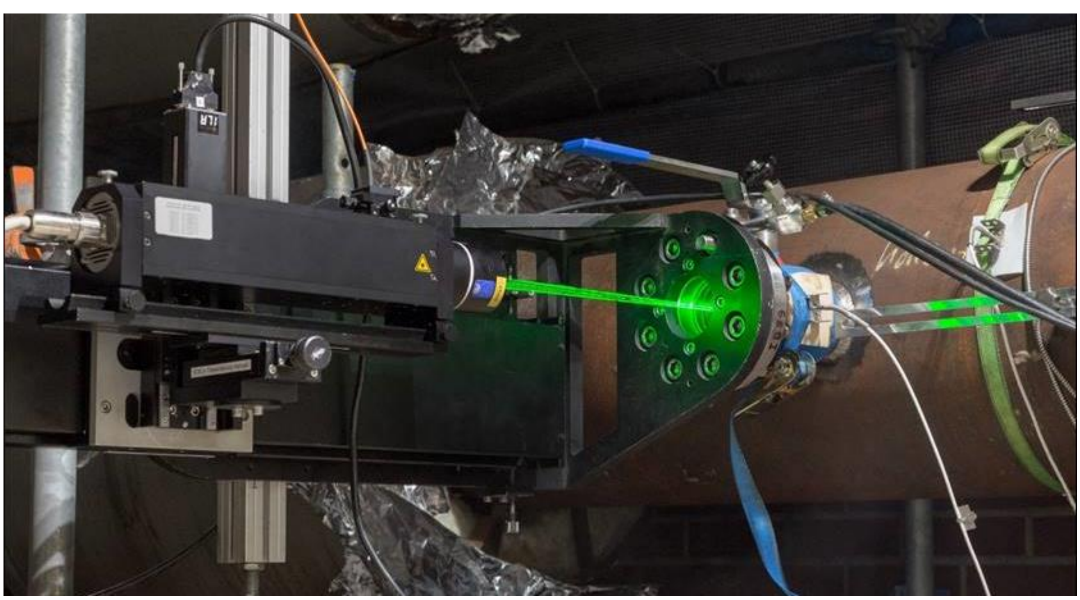


Figure 2. Photograph of the pipe with water flow and LDV on the traversing unit.

3. Temperature measurement using Brillouin scattering

If a liquid is irradiated with light, as in Figure 3, the molecules in the liquid scatter part of the light back, which is referred to as Brillouin scattering. Brillouin scattering occurs because the charges in the liquid molecules are excited to vibrate by the incoming electromagnetic light waves. Subsequently the molecules themselves emit light. This is a purely electromagnetic process without quantum effects. The spectral properties of the Brillouin scattered light depend strongly on the local speed of sound in the liquid, because the incident light waves interact with statistically occurring acoustic waves.

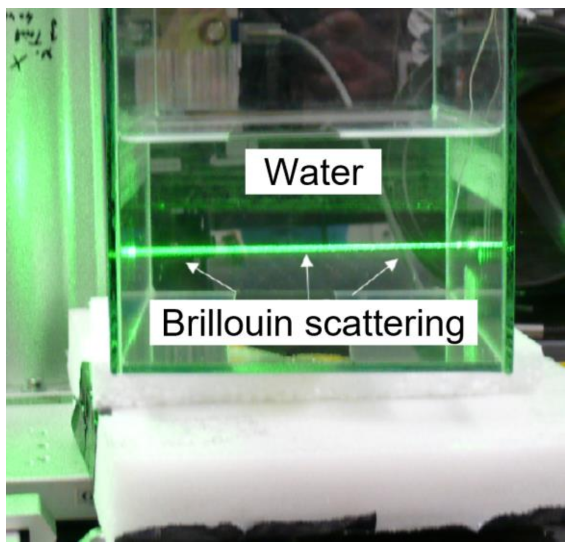


Figure 3. Brillouin scattering in water caused by a YAG laser beam (Enns, 2014).

In addition to Brillouin scattering, so-called Rayleigh scattering also occurs in liquids. It is also created by electromagnetic excitation but is independent of the speed of sound. Figure 4 shows how the proportions of Brillouin and Rayleigh scattering are distributed in the entire Rayleigh-Brillouin scatter spectrum. Brillouin scattering consists of two Brillouin profiles that are like Lorentz profiles and are shifted by the frequency $\pm f_b$ compared to the incident frequency f_0 of the excitation light. Rayleigh scattering is reflected in the spectrum by a profile centered around the incident frequency f_0.

Since the Brillouin scattering depends on the speed of sound and the speed of sound in turn depends on the local temperature in the liquid, the temperature can be derived from the shape of the Brillouin profiles. The precise calculation of the spectra is complex and as of today still subject of research. Some generally occurring effects are briefly listed below for clarification.

In addition to the speed of sound, the shift f_b also depends on the refractive index of the liquid, which also is temperature-dependent. In the case of water, the following relationship results approximately:

$$f_b = 2\frac{v_s\,n}{\lambda}\sin\frac{\theta}{2}, \qquad v_s = v_s(T,s)\,, \qquad n = n(T,s,\lambda).$$

Designations above are v_s: speed of sound, n: refractive index, λ: light wavelength, θ: angle between the direction of incidence and the direction of observation of the light, T: temperature, s: salt concentration of the water.

Figure 5 demonstrates how the frequency shift f_b depends on the temperature. The curve is also parameterized with the salinity. This should demonstrate that additives in the water can have an influence on the Rayleigh-Brillouin spectrum.

In addition to the parameters listed above, the width Δf_b of the Brillouin profile also depends on a quantity Γ, which measures the damping of density fluctuations. It is a function of various material properties of the water, of which not all are fully known (Fry et al., 2002):

$$\Delta f_b = \frac{\Gamma}{2}\left(\frac{4\pi n}{\lambda}\sin\frac{\theta}{2}\right)^2$$

The dependencies of f_b and Δf_b on the parameters listed above make it clear that the scattered light spectrum $S(f)$ of Brillouin scattering is a function of a large number of parameters:

$$S(f) = S(f, f_0, T, \theta, \lambda, \Gamma, s)$$

With regard to the planned temperature measurements, it is crucial that the scattered light spectrum S shows a pronounced temperature dependency.

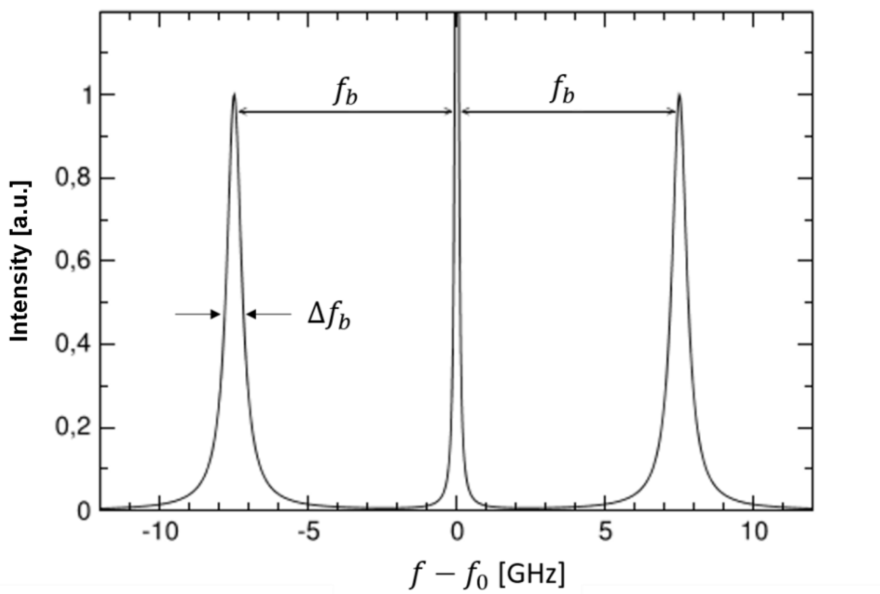


Figure 4. Calculated Brillouin spectrum in the case of visible light with frequency f_0. The red- and blue-shifted Brillouin lines are also referred to as Stokes or anti-Stokes components and result from the two possible directions of propagation of the sound wave in water. The central line is created by Rayleigh scattering in water (Schorstein, 2009).

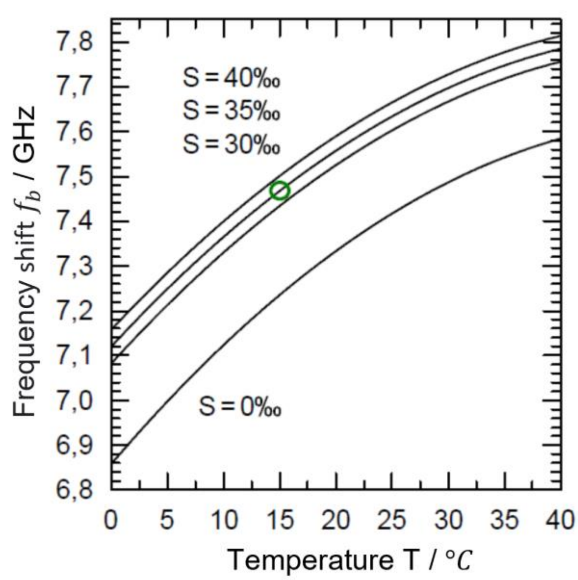


Figure 5. Change in frequency shift f_b with temperature T and salinity S of water (Rudolf, 2013). In the text, the salinity is denoted by S.

4. Modeling of the Brillouin spectrum

In order to be able to determine the temperature as precisely as possible using Brillouin scattering, a sufficiently accurate Brillouin model is required. So far, no model had been established to predict the width Δf_b of the Brillouin profile. Based on measurements by Fry et al. (2002), Popescu (2010) set up an empirical model for the profile width. However, in tests carried out by the project partner DLR, deviations of this model from the experiment have been shown, which are unlikely to result from measurement errors, see Figure 6. In particular, the decrease in profile width towards higher temperatures, which the Popescu model predicts, does not match the measurements carried out by Enns.

Various approaches were pursued in the project in order to improve the previous modelling. In the Popescu model, for an example, the influence of salinity is included by a linear approach, which probably is too simplified.

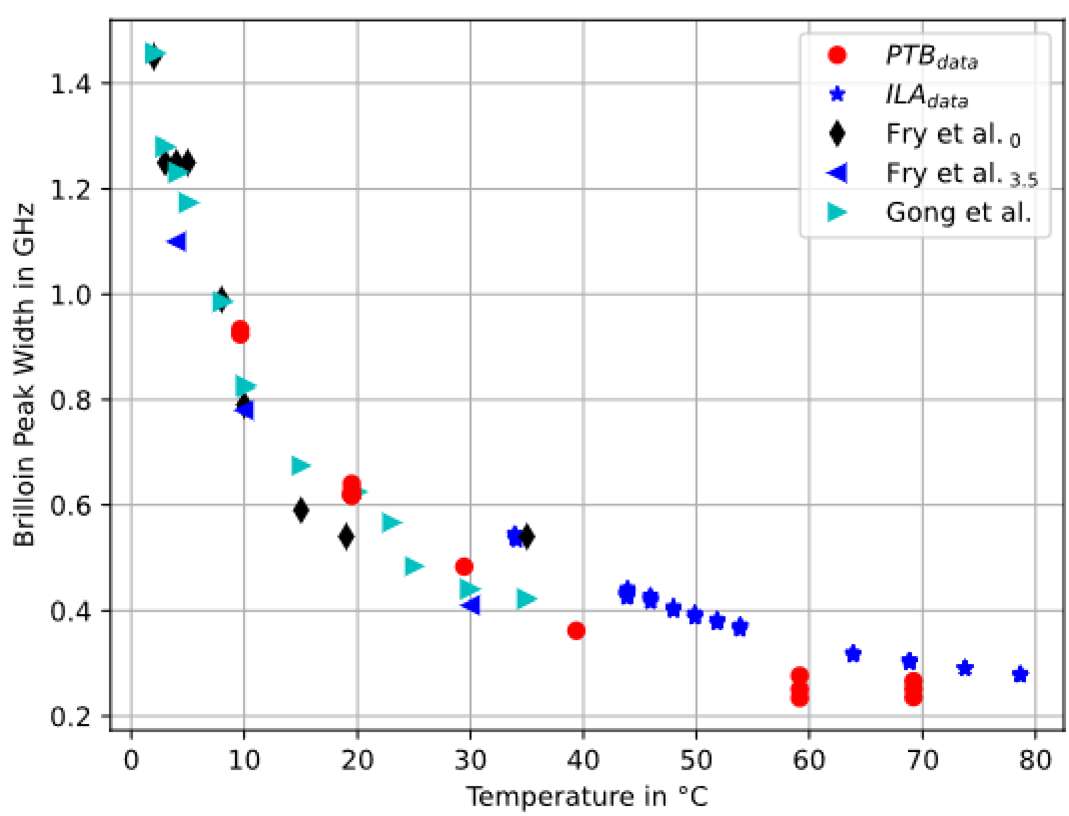


Figure 6. Brillouin peak width for water plotted against temperature. Red circles correspond to the measurements 2021 in the PTB. The blue stars were determined in the validation experiments at ILA R&D GmbH in 2021. Back diamond - low-salt water and left pointing triangle - salt water with a salinity of 3.5‰ according to Fry et al. (2002), turquoise right pointing triangle - according to Gong et.al. (2011).

5. Iodine filtering

Theoretically, it would be conceivable to sample the scattered spectrum of Rayleigh-Brillouin scattering *interferometrically* and to derive the desired temperature from the shape of the spectrum and the Brillouin model. In practice, however, such a procedure is ruled out because high-resolution spectrometers constructed from a resonator only have an extremely small optical aperture due to the principle involved. For a sufficiently strong measurement signal, however, scattered light must be collected from as large solid angle as possible.
The solution to this problem is to use the so-called *filtered* measurement technique. First of all, it must be taken into account that it is not even necessary to scan the spectrum exactly along a large number of frequencies, since in the end only one temperature value is to be derived from the entire spectrum. Another thing to keep in mind is that intensity measurements are much simpler than frequency measurements. These two considerations motivate the use of a *molecular absorption filter*, gaseous iodine in particular having useful properties and hence being often used.
In the practical version of the filtered measurement technology, the gaseous iodine is kept in a heated cuvette, the so-called iodine cell. This is placed in the optical path between the scattering area and the intensity detector, see Figure 7. The figure shows the exciting laser with the device for determining the laser frequency, the laser beam focal optics, the intensity detector with interchangeable lenses and the iodine cell.
The scattered light to be analyzed from the measurement volume is collected by the same lens that is used to illuminate the measurement volume and imaged through the iodine cell onto the intensity detector. The absorption in the iodine vapor causes a characteristic weakening of the scattered light, which can be described mathematically by the product of the transmission $\tau(f)$ of the iodine vapor and the scattered light power $S(f, f_0, T)$. The intensity detector, which sensivity is not frequency-dependent in the relevant frequency range integrates the product of both spectra so that the resulting intensity is:

$$I = I_0 \, R \int \tau(f) \, S(f, f_0, T) \, df \qquad (1),$$

I_0 is the power of the laser. The quantity R describes the optical sensitivity (or efficiency) of the setup. It includes all device-specific attenuation factors, such as scattering on lens surfaces and

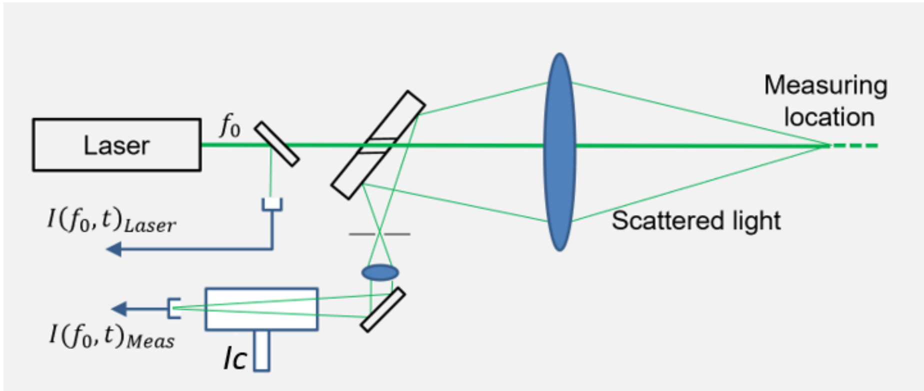


Figure 7. Basic structure of a system for temperature measurement using filtered Brillouin scattering.

also possible absorption in turbid water. It is assumed that R is not a function of the scattered light frequency because the scattered light spectrum is very narrow.

Instead of resolving the entire scattered light spectrum, only a *single* intensity value is measured for the filtered measurement technique. To still be able to derive meaningful statements from the measured intensity value despite this extreme reduction in information, the special absorption properties of iodine vapour are used, consisting of many absorption bands with steep slopes. Figure 8 shows the iodine transmission τ in a selected, narrow wavenumber range. Three minima can be seen in the transmission, which correspond to three absorption bands.

The three transmission minima were chosen such that the exciting laser frequency corresponds to the middle minima and the red- and blue-shifted Brillouin profiles correspond to the other two minima. If the water temperature varies, the position and width of the Brillouin profiles change and the integral according to (1) leads to a different intensity reading of the detector. The steep slopes of the iodine transmission curve near the minima mean that even small changes in the position and width of the scattering spectra lead to a strong variation in intensity. In this way, the iodine filter acts as a sensitive "frequency-to-intensity converter" that converts hard-to-measure frequency changes into easy-to-measure intensity changes.

The problem of a small optical aperture when using a spectrometer was discussed above. This problem does not exist with the filtered measurement technology since the diverging scattered light from the measurement volume can be bundled using lenses and guided through the iodine cell without any problems. A high signal yield is obtained in this way.

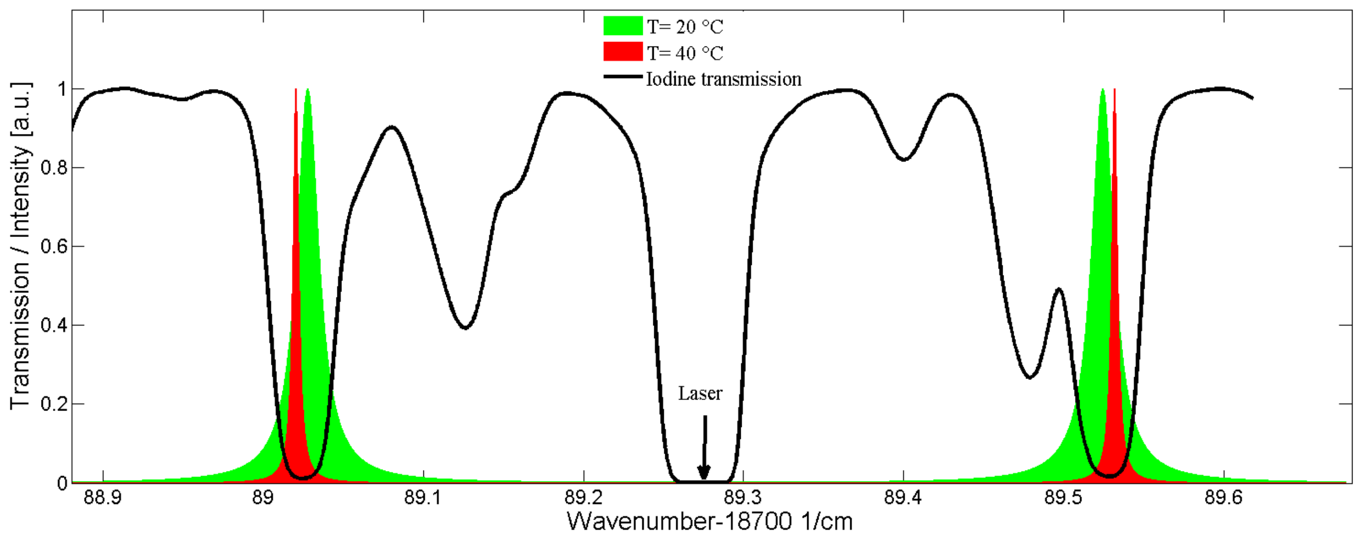


Figure 8. Iodine transmission curve and Brillouin spectra (colored) of water for two temperatures. The Rayleigh spectrum is not shown. Source: DLR.

The use of the iodine filter has the second advantage: if the excitation frequency of the laser is selected so that it falls within a transmission minimum of the iodine cell, as in Figure 8, all undesired forms of scattered light are prevented from reaching the detector. This applies in particular to light that is scattered by particles (Mie scattering) which is several magnitudes stronger than Brillouin scattering. Every water flow contains particles to a certain extent, most of which produce a relatively strong scattered light signal, that would severely disturb the Brillouin measurement if no countermeasures are taken. However, these particles are anecessity for the LDV technology discussed below. Determining the temperature using Brillouin scattering therefore requires the suppression of scattered light from particles. This is what the iodine cell can achieve because the Mie scattering is only very slightly shifted compared to the excitation frequency due to the Doppler effect. It therefore still falls within the central transmission minimum and is almost completely absorbed by the iodine cell.

Reflections of the laser light on surfaces such as windows or walls must also be suppressed. This strong stray light is blocked by the iodine cell as well because its frequency is the same the excitation frequency. This enables measurements close to the wall, which is very useful both for accurate measurements of heat flow and for studies of heat transfer at a wall.

6. Quotient method and frequency scanning method

In order to be able to determine the temperature from measured intensities using (1), one needs a Brillouin model that establishes the relationship between the temperature and the scattered

spectrum. Additionally, the optical sensitivity R must be determined. There are two possible methods for this: the quotient method and the frequency scanning method.
With the quotient method, the optical sensitivity of the setup is determined from a reference measurement. The detector's intensity is measured for a liquid of known temperature. In this way one can use (1) to determine the optical sensitivity. The name "quotient method" originates from the fact that one can eliminate the factor R in (1) by dividing the equation for an unknown temperature by the same equation at the known reference temperature and reference intensity.
The quotient method has the advantage that it is fast. Its disadvantage, however, is the additional reference measurement that is required. A reference measurement can be avoided if the necessary information for determining the optical sensitivity is accessed in a different way. This is what the frequency scanning method does.
During the scanning process, the exciting laser frequency f_0 is changed slightly step by step and the intensity transmitted through the iodine cell is measured in each case. The intensity changes for the following reason: If the excitation frequency is shifted, the center frequency of the Brillouin profiles also shifts by the same amount (compare Figure 4). Therefore, they assume a different position relative to the iodine transmission curve, and a different amount of Brillouin scattered light passes through the iodine cell. The combination of the frequency shift of the scatter profile and the integration according to (1) can also be understood as a convolution between the scatter spectrum and the iodine transmission.
An example result of the frequency scanning method is shown in Figure 9. A characteristic parabolic progression of the intensity can be seen for all temperatures with changing excitation frequency (or wave number). The occurrence of a minimum can be explained by the fact that the two Brillouin profiles in Figure 8 optimally fall into the two transmission minima at the corresponding excitation frequency, so that absorption is maximal.
With the frequency scanning method, the amount of information available is increased compared to the quotient method, so that there is not one intensity at one excitation frequency, but rather a set of intensities for a set of excitation frequencies. This turns the individual equation (1) into a set of equations, i.e. a system of equations. This system of equations provides enough information to determine the two quantities R and T that are being sought. If more than two excitation frequencies are used, the system of equations is overdetermined. Due to measurement and modeling errors, not all equations can be fulfilled exactly in such a case; and it makes sense to look for a solution where all equations have the smallest error on average. An iterative solver can be used to solve such a system of equations. In the current ZIM project on the subject of DGV/FRS, the Levenberg-Marquardt method has proven to be suitable in this respect.

The sampling method has the advantage that it does not require the reference measurement required for the quotient method. Another advantage is that stochastic and systematic errors that occur at the different excitation frequencies are averaged out, so that the measurement accuracy is increased. The disadvantage of the sampling method is the longer measurement time.

7. Experimental set-up and results

For the FBS method, a cw single mode laser is detuned in its frequency step by step, the scattered light is collected from a laser focus and the transmission of this scattered light through a molecular absorber, a so-called iodine cell, is measured. A typical setup is shown below in Figure 10.
The scattering process and the characteristic shape of the scattered light's spectrum can be described with suitable models, so that the temperature as well as other parameters of the flow can be derived by comparing measurement and model.
By combining the optical temperature measurement system with an established method to measure the volumetric flow profile (in this case by LDV), a laser-optical heat flow sensor is created. This could for example be used to study the heat flow in district heating or cooling systems or other applications with hot or cold water flows.
To achieve measurement results under very defined conditions, a special flow test facility was created by the Physikalisch-Technische Bundesanstalt (PTB) (see Figure 11). The flow facility either provides a water flow of very stable temperature or alternatively can generate rapid temperature fluctuations. Additionally, also spatial temperature profiles and stratification can be demonstrated. Measurements were carried out at this facility with the FBS system designed and built by ILA R&D GmbH (Figure 10).

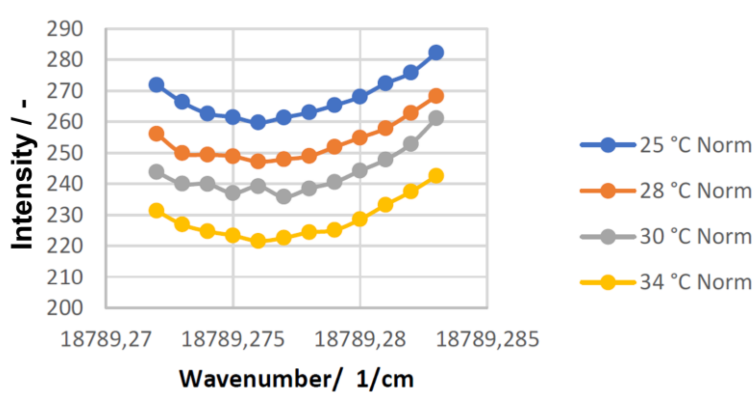


Figure 9. Measured intensities at different excitation wavenumbers and water temperatures.

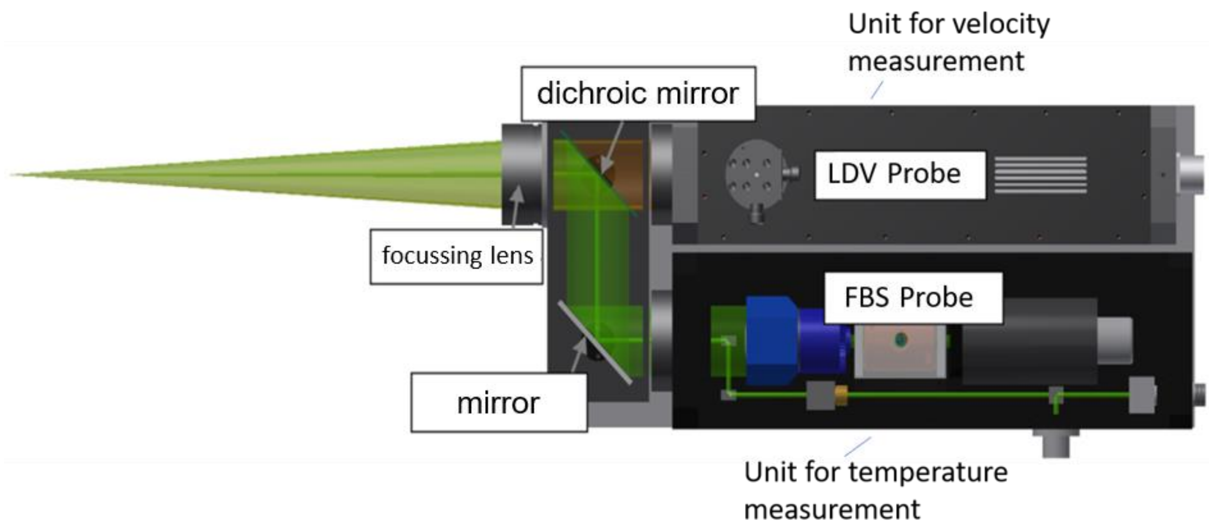


Figure 10. Sectional drawing of the combined LDA-FBS probe

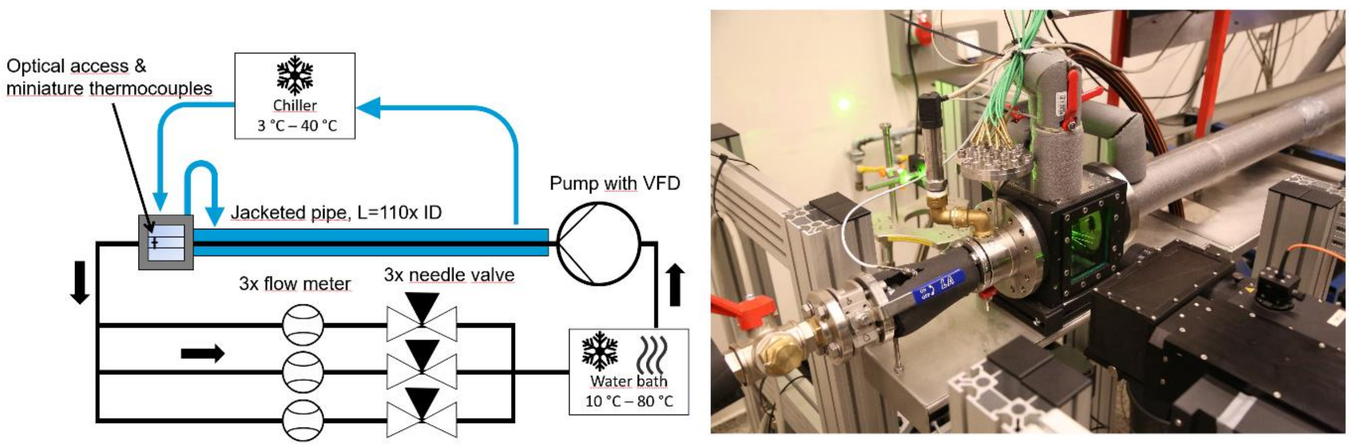


Figure 11. Flow diagram of the flow test bench with temperature measuring points (left). Photograph of Flow test bench at PTB (right)

Figure 12 shows FBS spectra at different water temperatures. Figure 13 compares temperatures measured by both FBS and thermocouples. The measurement accuracy turned out to be mostly better than $\pm 1\,°C$, which is in accordance with theoretical considerations.

It is crucial to use a proper model of the scattering process to achieve accurate measurement results. The models found in the literature have been improved as part of this project, as they turned out to be not precise enough and additionally show a dependency on water properties like salinity and conductivity. In particular, it was possible to determine the line width of the individual Brillouin peaks more precisely than previously published in the literature. In this respect, the project also proved important for basic research.

8. Temperature measurement with high temporal resolution

Figure 14 shows the measured absolute temperature compared to the reference temperature measured by a PT100-4W sensor and Figure 15 the comparison between the FBS temperature signal and the thermocouples. Three different methods were used for this. In the first method (Brillouin width), the entire intensity curves were compared with the FBS model and the temperatures were determined through the model. With the other two methods, only a single intensity value at one laser wavelength at the minimum of the intensity curves was compared with the model (single wavelength), or the intensity values at two wavelengths (dual wavelength). By determining only one or two intensities, the measurement time is reduced, which is particularly important for transient temperature measurements.

The results show that the temperature determination displays an offset over the entire Brillouin intensity curve and is significantly imprecise above 43 °C. One conjecture is that these phenomena can be resolved by improving the FBS model. The measurements at only one or at two wavelengths showed good agreement with the reference temperatures over the entire measured temperature range and a mean deviation of less than 1 K.

Using the method of continuous frequency detuning and the simultaneous recording of the filtered scattered light intensity, the time-averaged mean values of the water temperature can be determined with good precision. In order to also measure rapid temperature fluctuations with a temporal resolution of up to 100 Hz, another measuring method is used simultaneously: the intensity of the scattered light after it has passed the absorber cell is also a function of the temperature. This method is primarily characterized by the fact that a temperature difference can be determined fast and with good precision.

However, this method does not yield good absolute temperature values. It was therefore combined with the frequency scanning method. This provides a way to establish an absolute temperature value with good accuracy and determine the relative changes of the temperature in relation to this value with high temporal resolution (Figure 15).

Figure 15 shows the course of such a rapid temperature transient. On the one hand, one can see the temperatures measured by the thermocouple. The second diagram shows the temperature profile measured with the FBS method. This shows a clearly faster increase, which indicates a higher cut-off frequency of the optical method. Particularly impressive is the small, pointed peak at the beginning of the temperature rise, which is a special effect of the fluid mechanics equipment used and which can be found very nicely in both measurements.

The long-term goal of this development is to bring a device into the market with which the heat flow can be determined by combining the flow measurement using LDV with temperature measurement using FBS. The time-resolved temperature measurement is of particular importance for this purpose, as it would enable to determine a correct mean value of the heat flow even in a flow with fluctuating temperature and velocity or with a significant spatial profile of those quantities over the flow cross section.

Since the line shape of the Brillouin spectrum depends on the water composition, it is important that the user can calibrate the measuring device with the water of his experiment. For this purpose, OPTOLUTION developed a special calibration device, which is shown Figure 16.

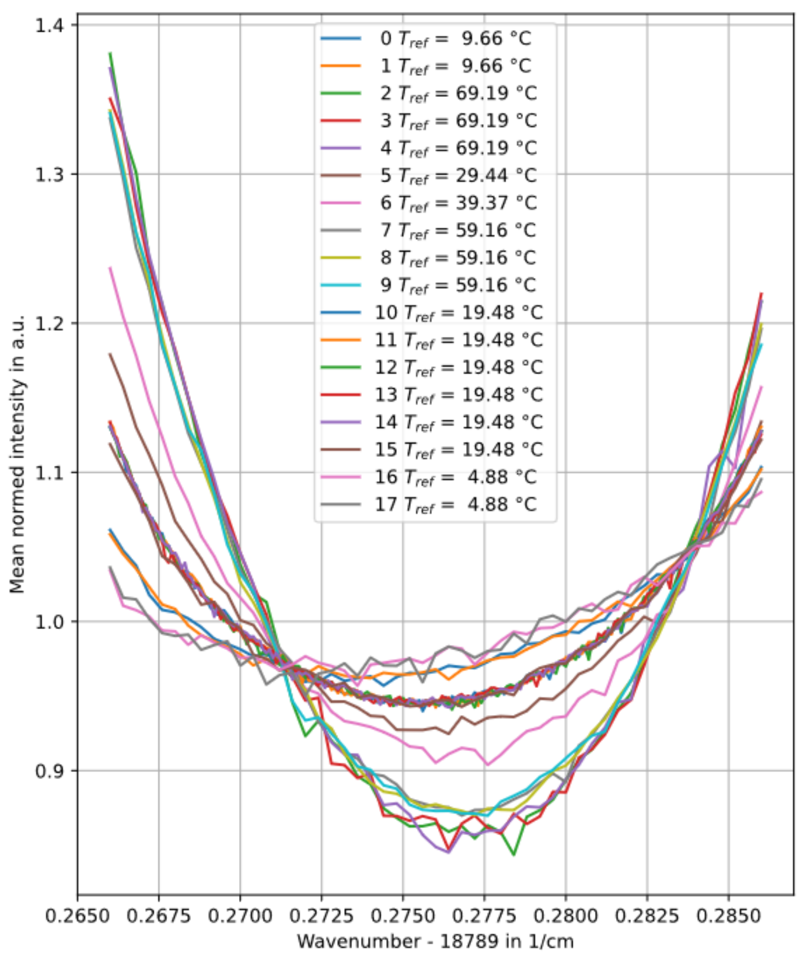


Figure 12. Frequency scan measurements for stationary conditions. The normalized intensity of the filtered scattered light over the wave number is shown. With temperature increase, the parabola-shaped curve becomes wider.

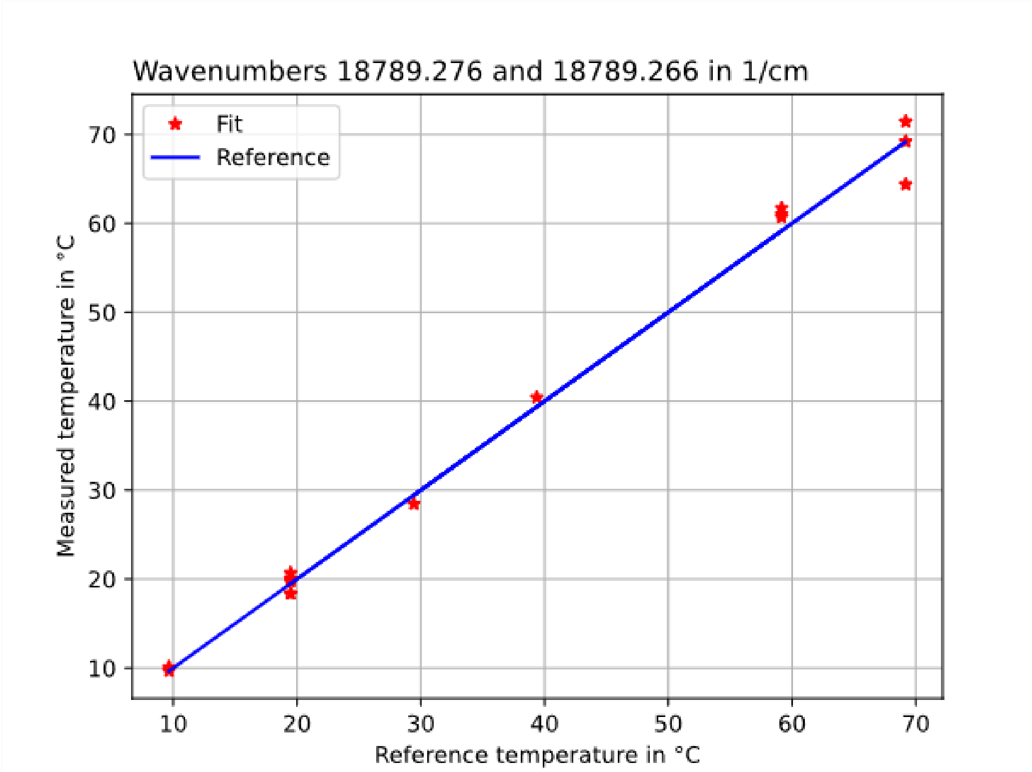


Figure 13. FBS temperature plotted against the reference temperature using the dual wave number method.

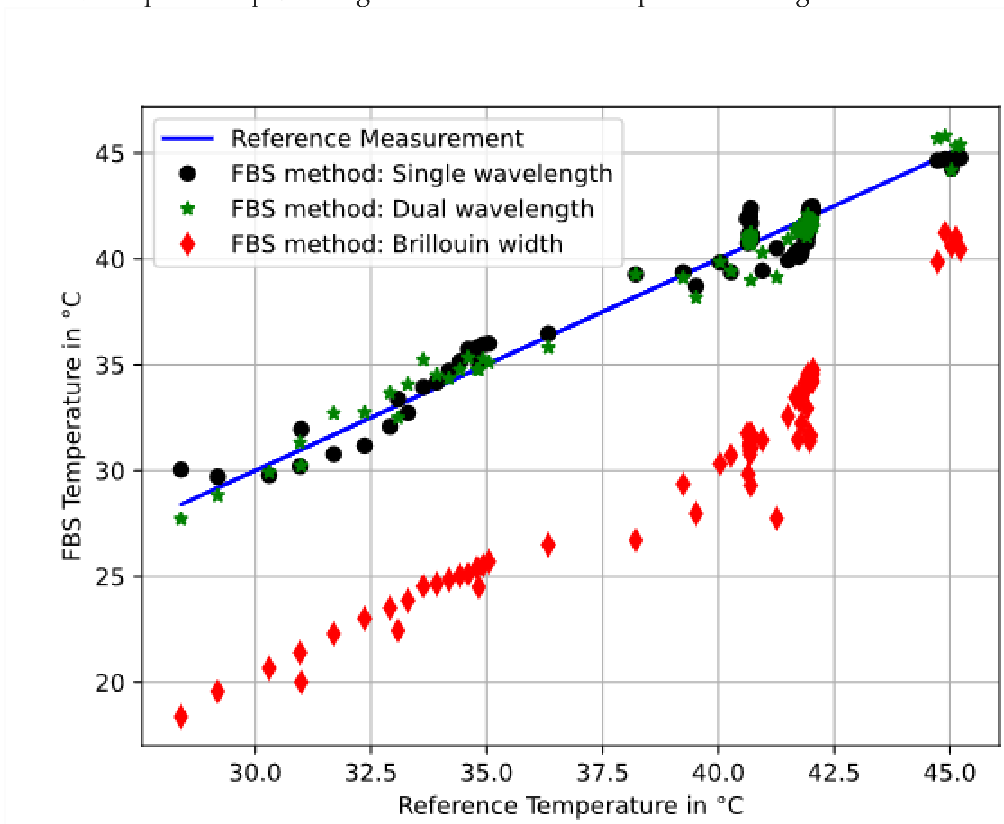


Figure 14. Comparison of the temperatures measured by the FBS probe with the reference temperatures of a PT100 temperature sensor

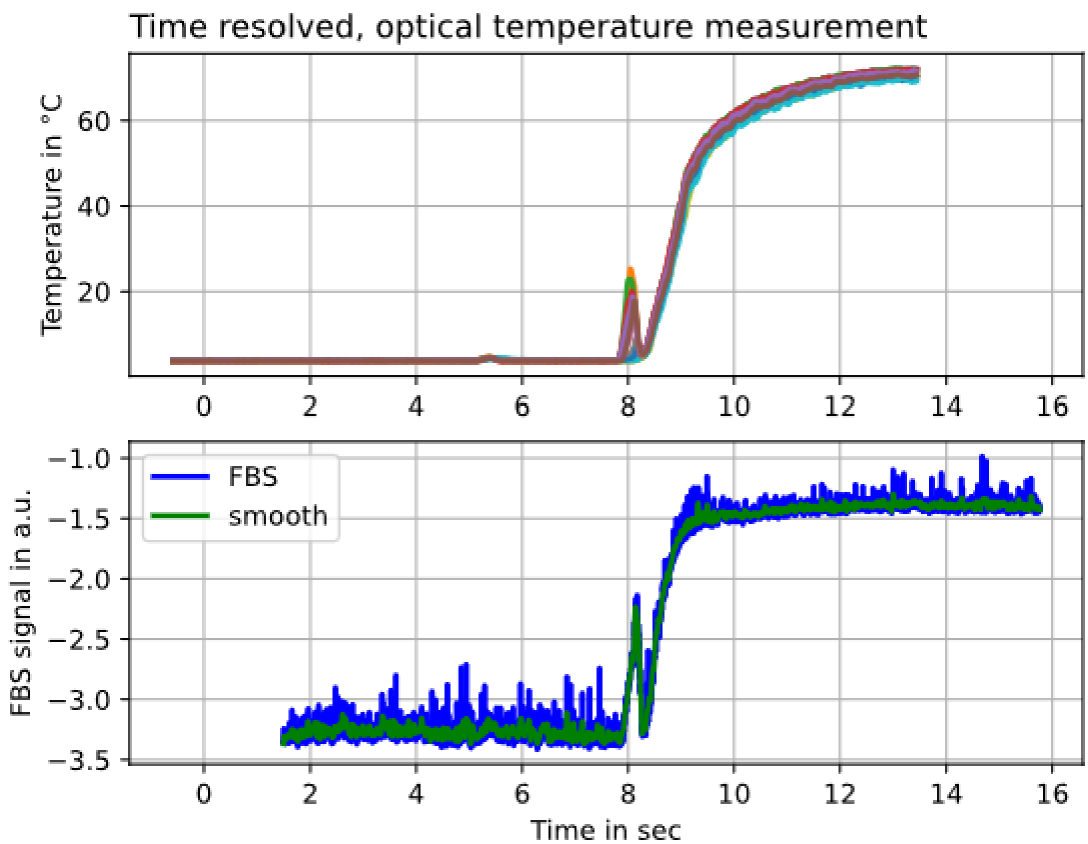


Figure 15. Time resolved, optical temperature measurement. The reference temperature is plotted in degrees Celsius over the measurement time. The different colors each correspond to one of the 16 temperature sensors of the reference measurement. The normalized FBS signal (in arbitrary units) over time is shown below (blue). With a smoothing filter, the course shown in green is obtained.

9. Conclusion and outlook

Basic studies on system design and on the influence of particle scattering were carried out by DLR. The FBS data was evaluated using the VIPER software, which was developed by BHT. On the one hand, various models of the scattering process are implemented in this software and, on the other hand, an especially well-suited optimizer that utilizes the Levenberg-Marquardt algorithm. The temperature measurement system must be calibrated using a water sample of the fluid to be measured. For this purpose, OPTOLUTION GmbH has developed a custom-built, very precise calibration device (Figure 16).

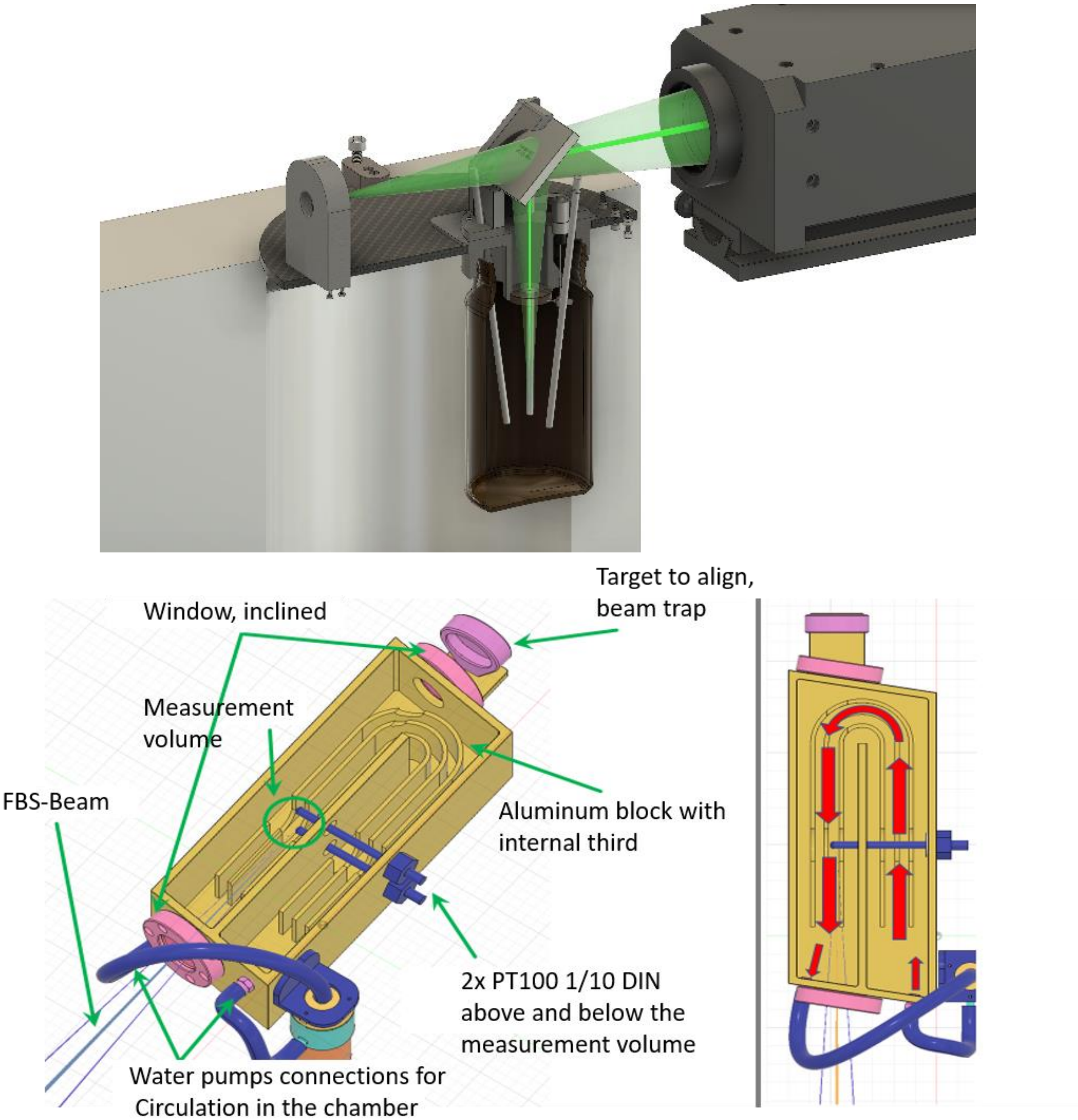


Figure 16. Two versions of a custom-built calibration device.

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