Thermographic laser Doppler velocimetry using the phase-shifted luminescence of BAM:Eu²⁺ phosphor particles for thermometry

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Abstract: Simultaneous point measurements of gas velocity and temperature were recently demonstrated using thermographic phosphors as tracer particles. There, continuous wave (CW) excitation was used and the spectral shift of the luminescence was detected with a two-colour intensity ratio method to determine the gas temperature. The conventional laser Doppler velocimetry (LDV) technique was employed for velocimetry. In this paper, an alternative approach to the gas temperature measurements is presented, which is instead based on the temperature-dependence of the luminescence lifetime. The phase-shift between the luminescence signal and time-modulated excitation light is evaluated for single $BaMgAl_{10}O_{17}:Eu^{2+}$ phosphor particles as they cross the probe volume. Luminescence lifetimes evaluated in the time domain and frequency domain indicate that in these experiments interferences from in-phase signals such as stray excitation laser light are negligible. The dependence of the phase-shift on flow temperature is characterised. In the temperature sensitive range above 700 K, precise gas temperature measurements can be obtained (8.6 K at 840 K) with this approach.

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1.   Introduction

Thermographic phosphors may be used as flow tracers for simultaneous temperature and velocity measurements in fluids. While flow velocity measurements using these particles are accomplished by the conventional use of the Mie scattered laser light as in the particle image velocimetry (PIV) or in laser Doppler velocimetry (LDV) techniques, phosphor thermometry exploits the temperature dependence of the luminescence properties of the particles, by detecting the emission following laser excitation [1-6]. If micrometer-size particles are used, they rapidly respond to changes in fluid temperature and flow velocity (< 100 μs in air at 800 K [3]). To infer the particle temperature, two common methods can be used: the two-colour intensity ratio approach; or the lifetime decay approach. The intensity ratio approach, in which the ratio of two spectral regions of the emission spectrum is temperature dependent, has been used extensively for planar excitation flow temperature measurements e.g. [1-5]. Also we have recently demonstrated a point measurement technique for flow temperature and velocity measurements, which is based on detecting the luminescence and Mie scattering signals of individual phosphor particles crossing continuous wave (CW) laser beams using photomultiplier tubes (PMTs) [6]. For the temperature measurement the luminescence emission, which is time-integrated over the particle transit time through the probe volume, is spectrally filtered in two bands and divided to generate the intensity ratio. This "thermographic LDV" technique offers point measurements of flow temperature and velocity at a fast sampling rate (kHz) and with a high spatial resolution (~150 μm). These latter capabilities greatly advance the utility of correlated vector-scalar statistics, which are important to characterise turbulent heat transport.

For measurements on static surfaces, the lifetime-based approach was shown to offer, in general, improved accuracy, precision, and was shown to be less sensitive to optical alignment in comparison to the spectral methods [7]. However, for measurements in turbulent flows, normally the measurement duration is limited to 10's microseconds, which then requires both a detector with a high sampling rate (10's MHz), and a phosphor with a fast (1-10 μs) luminescence lifetime that is sensitive in the desired temperature range.

There are reports on the use of the phosphor La$_2$O$_2$S:Eu^{3+} for the measurement of temperatures of liquid films, droplet and sprays [8-10]. These studies used low repetition-rate pulsed nitrogen or Nd:YAG lasers for excitation, and PMTs or ultra-high-speed framing cameras (~100 MHz) for point and 2D detection respectively. The lifetime was evaluated in the

time domain after laser excitation. The $^5D_2$ line of $La_2O_2S:Eu^{3+}$ has a lifetime of 12 μs at room temperature affording short overall integration times, and covering the temperature range up to ~400 K.

The current work combines the advantages of high sampling rate afforded by the above-mentioned thermographic LDV technique and the extraordinary temperature sensitivity of the luminescence lifetime. This new approach is based on the phase shift of the luminescence of single phosphor particles relative to the time-modulated CW UV excitation laser light. The luminescence emission and Mie scattering of the excitation light, detected during the transit of each seeded particle through the focused laser beam, oscillate at the frequency of the modulated laser source. The lifetime of the luminescence transitions results in a phase shift of the emission signal relative to the scattered excitation light from which the particle temperature can be determined. The same particles are used to determine the velocity as they cross a set of fringes overlapping the probe volume, generated by crossed green laser beams as in ordinary LDV. The following expands on the phase shift concept and the phosphor used in this study

2.   Phase shift concept and the thermographic phosphor BAM:Eu^{2+}

Unlike luminescence lifetime measurements, which typically implement pulsed light sources, phase shift measurements require the use of a modulated excitation source. The intensity of the incident excitation light is varied at a high frequency, in the range of the reciprocal of the phosphor luminescence decay time (or 'lifetime') τ. The luminescence signal following this excitation mode responds at the same modulation frequency, but is delayed due to the lifetime of the luminescence transition, thereby resulting in a phase shift of the emission signal with respect to the excitation light, as shown on Fig. 1. For a mono-exponential decay, the relationship between the phase shift ($\Delta\Phi$) and the decay time (τ) is given by Eq. (1) [11];

$$\tan(\Delta\Phi) = 2\pi f_M \tau \tag{1}$$

where f_M is the modulation frequency of the oscillating excitation light intensity. Measuring the phase shift therefore exploits the temperature dependence of the lifetime. When an arbitrary waveform is chosen to drive the excitation source, thereby producing light intensity in an oscillating manner, both luminescence and excitation signals can be cast in the Fourier space to determine their frequency and phase contents.
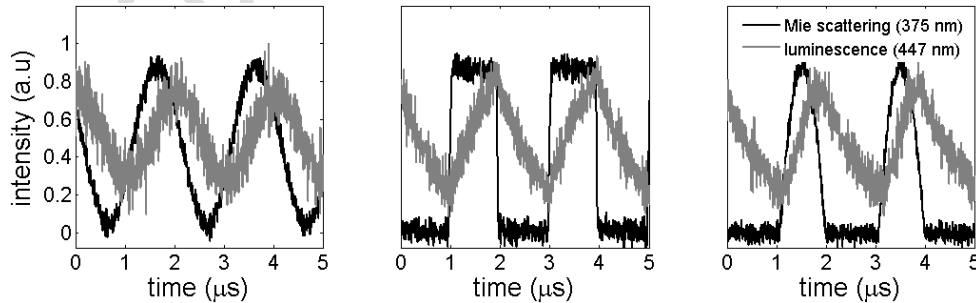

Fig. 1. Luminescence and Mie scattering signals from three different excitation waveform schemes (a) 0.5 MHz sine wave, (b) 0.5 MHz square wave, and (c) 0.5 MHz half sine wave recorded from a stack of aggregated BAM:Eu^{2+} phosphor powder. The luminescence signal is collected using a bandpass filter 447-60nm.

Here, in the context of flow measurements, as a particle crosses the beam with a Gaussian intensity profile, the temporal evolution of the excitation intensity has a Gaussian envelope,

with a width that is linked to the particle transit time through the probe volume. Since the Mie scattering from the particles at the UV excitation wavelength reflects exactly this excitation waveform, this signal is detected and used as the reference for the phase shift evaluation.

To demonstrate the feasibility of the proposed approach for the application of phosphor thermometry to flow diagnostics, a phosphor with adequate luminescence properties in term of brightness (emission intensity), lifetime and temperature sensitive range must be chosen. In this work, the phosphor europium doped barium magnesium aluminate ($(BaMgAl_{10}O_{17}:Eu^{2+})$, hereafter referred to as $BAM:Eu^{2+}$) was used. The luminescence characteristics were studied extensively in the gas phase [12] and the temperature dependence of the lifetime [13] and the emission spectrum [12] have been reported. $BAM:Eu^{2+}$ is readily excited by a 375 nm CW UV laser diode (also employed in the previous thermographic LDV work [6]) and has a high the quantum yield (> 90%), which is important because single particles are probed in the gas which normally limits signal levels. A mono-exponential curve, Eq. (2), may be used to fit the experimental data of the luminescence decay, as described by;

$$I = I_0 \exp(-t / \tau) \tag{2}$$

where I is the luminescence intensity, I_0 is the luminescence intensity at t = 0, t is the time after termination of the excitation pulse and τ is the luminescence lifetime. The lifetime of $BAM:Eu^{2+}$ is around 1 µs at room temperature. Also, the lifetime strongly decreases at temperatures above 600 K [13] due to the increasing non-radiative transition rate. For simultaneous measurements, the peak wavelength of the emission spectrum of $BAM:Eu^{2+}$ is around 445 nm, and can easily be spectrally filtered from both Mie scattering light from incident UV excitation (here 375 nm) and that of the visible light, for example 514.5 nm, argon-ion laser, which is required for the LDV measurements. Thus, an optical filter with central wavelength around this 445 nm peak can be used to collect nearly all of the luminescence from $BAM:Eu^{2+}$.

Two separate studies have shown that the oxygen concentration (in the investigated range from 0 to 200 mbar of O_2 partial pressure and up to 920 K) [12], or the pressure (up to 10 kbar, at room temperature) [14] has no noticeable effect on the luminescence emission. For a given phosphor material, changes in the luminescence lifetime are mainly the result of a change in the non-radiative transition rate, so they must be associated with changes in quantum yield, and therefore changes in emission intensity. Since no changes in emission intensity were observed under the different environments in the cited studies, it is unlikely that changes in luminescence lifetime occur. In principle then, no cross-sensitivity of the phase-shift to pressure and/or oxygen concentration are expected. For these reasons, $BAM:Eu^{2+}$ possesses appropriate luminescence properties for applying the phase-shift concept in turbulent gas flows to measure above 600 K.

The following describes the experimental setup of the thermographic LDV thermometer where the phase shift is used to determine the fluid temperature, including the data processing, and the test rig to assess the measurement concept. The temperature precision is evaluated in the gas phase. Additional measurements are performed to assess the possible contribution of in-phase interfering signals generated by the excitation laser. The main conclusions are summarised in Section 5.

3. Experimental Set-up

3.1 Gas-phase measurements

The experimental set-up was similar to that used previously for ratio-based point measurements [6] but with a few changes to the optical components and arrangement. An air jet was

electrically heated using temperature-controlled heaters (1245 W heating tape, Omega Engineering) wrapped around an 8 mm internal diameter circular tube. 2.4 µm diameter BAM:Eu^{2+} particles (KEMK63/UF-P1, Phosphor Technology) were seeded into the flow using a magnetic stirrer-type seeder. The temperature and velocity 95% response times are both around 50-100 µs respectively, assuming 2.4 µm spherical phosphor particles in air at 800 K, using the approaches described in [3].

As in [6], the LDV measurement volume was formed at the intersection of two crossed 514.5 nm laser beams from an argon ion laser (BeamLok 2060, Spectra Physics) as shown in Fig. 2. To excite the phosphor particles, a third beam from a 70 mW UV (375 nm) diode laser (PhoxX, Omicron Lasers) was superimposed on the LDV measurement volume using the same focusing lens. Using a beam sampler, a small fraction of the three beams was directed onto a CCD to measure the size of the three beams (all ~150 µm e^{-2} diameter) and monitor their overlap.

The UV laser was modulated using an arbitrary waveform generator (TGA1241-Thurlby Thandar Instruments). Three waveforms were considered: a sinusoid, referred to FSW (Full sine wave), a sinusoid truncated to its upper half (hereafter called the half sine wave), and a square wave. Examples of such excitation waveforms were shown in Fig. 1. For application of the phase shift concept to flow thermometry, where single phosphor particles crossing the finite probe volume (~150 µm diameter) with various flow velocities are probed, a sufficiently high modulation frequency is required. This is important because the number of cycles of the modulated incident light which a particle is exposed to depends on its transit time (velocity) through the probe volume. Here, the 1 MHz modulation frequency resulted in about 75 cycles at a velocity of 2 m/s.

Optical signals from the combined probe volume were collected perpendicularly to the axis of beam propagation by a single 85 mm (f/1.4D) Nikon camera lens. A 200 µm slit placed at the focus of the lens in the image plane was used to define the extent in the horizontal direction of the probe volume in the object plane. The signals were collimated by a plano–convex lens, f = 75 mm, and separated by a long-pass dichroic beam splitter (Chroma-t500lpxrxt-UF1), which transmits the 514.5 nm Mie-scattering (Doppler bursts) towards PMT1, equipped with a 514-10 nm (notation is CWL – FWHM) bandpass filter and reflects the 375 nm Mie-scattering and luminescence bursts. These signal were separated using a combination of a 50:50 plate beamsplitter (BSW10R, Thorlabs) and a pair of interference filters; 375–10 nm (OD $\geq$ 4) PMT2 and 447–60 nm (OD $\geq$ 6) PMT3 (86736 and 84095, Edmund Optics). A 400 nm long-pass colour glass filter (Schott GG400) was also used on the luminescence channel (PMT 3), to remove any remaining scattered 375 nm light.

The filtered signals were focused onto the PMTs (PMT1; (PMTSS, Thorlabs), PMTs 2&3 (R955HA, Hamamatsu)) using 50 mm lenses. Each PMT was terminated by a 100 Ω resistance and the signals were digitized by a DPO4054 oscilloscope (Tektronix). The supply voltage of of PMT 2 & 3 was set to 395 V and 570 V respectively. With the luminescence signal set as the trigger source channel, individual burst signals surpassing a set trigger level (560 µV) were saved, and the acquisition was sustained for 100 repeated particle crossings across the probe volume for each measurement set. The phase of the 375 nm Mie scattering signal was used as reference relative to the phase of the luminescence signal, that is, evaluating the PMT 2 and 3 signals using a fast Fourier transform (FFT) algorithm to determine the phase shift,

$$\Delta\Phi = \Phi_{PMT2(375nm)} - \Phi_{PMT3(447nm)}$$ each time a particle crosses the probe volume.

Simultaneously, the 514.5 nm Mie scattered light from the same particles crossing the fringes like in the conventional LDV was used to evaluate the flow velocity, also using an FFT algorithm, to extract the Doppler frequency.
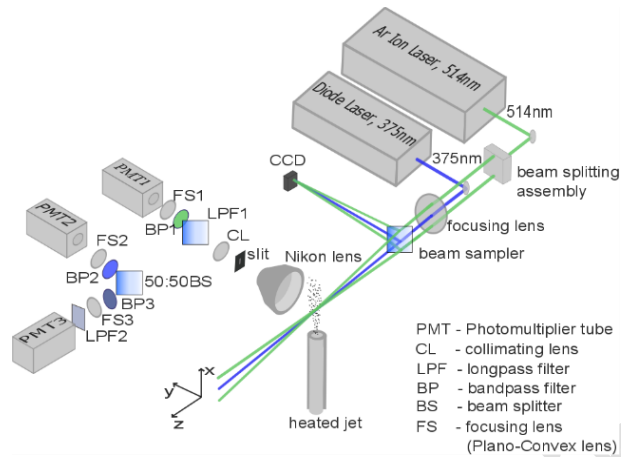
Fig. 2. Experimental setup for simultaneous flow temperature and velocity measurements. LPF1: 500 nm, LPF2: 400 nm, 50:50BS: plate beam splitter, BP1: 514-10 nm, BP2: 375-10 nm, BP3: 447-60 nm, FS1, 2, and 3: focal length = 50 mm.

3.2 Investigation of in-phase signals

For time-domain luminescence lifetime measurements, the decay time can be evaluated over a time window when the excitation light source is off, so that interferences from fast fluorescence and scattered laser light are minimised. This temporal separation is not possible in the phase shift concept. Depending on the intensity and spectral position of those signals, the optical density of the interference filters, and the rejection of stray light, these signals may not be fully suppressed. Such interference may alter the measured phase shift. It is therefore important to investigate a potential contribution of non-phase shifted light to the measurements. In part, this can be achieved by comparing the phase-shift measured using the modulated excitation source to the lifetime evaluated from luminescence decay waveforms recorded for individual excitation pulses.

Various excitation waveforms were employed as permitted by the diode laser, to investigate the potential interference from in-phase signals on phase shift measurements. Single pulses and square wave modulation permits evaluation of the decay time in the temporal domain over time windows where the laser is off, avoiding influence from fast fluorescence and scattering. Square wave modulation, in particular, allows the evaluation of both decay time and phase shift using an identical excitation waveform. For this part of the study, the optical setup shown in Fig. 2 was employed, but with both the argon ion laser and PMT1 turned off, and with slight modifications made to the setup.

To avoid the Gaussian modulation resulting from the particle transit through the laser beam, the seeded flow was replaced by a static stack of aggregated powder of the same BAM:Eu^{2+} phosphor. Here, all the measurements were carried out at room temperature. To monitor the excitation light, a beam sampler was used to direct a portion of the beam toward a photodiode (DET10A, Thorlabs), which replaced the CCD. For these tests, by calibrating the photodiode with the optical intensity of the laser under CW mode operation, the maximum output optical power for each pulse (67.8 mW) was monitored and kept constant for the three waveform schemes (half sine wave, full sine wave and square wave). Modulation frequencies from 50 kHz up to 1 MHz were employed. For each waveform and at each modulation frequency, 20 measurement sets were taken.

In the powder-based measurements, initially a lower PMT supply voltage was used (224 V) because the luminescence signal is higher than in the gas-phase. This may influence the temporal response of the detection system. Thus for comparison, an experiment was also

performed with the PMT supply voltage set to 570 V, the same value as the gas-phase measurement. In that case, a 0.1% transmission neutral density filter was used to operate on the same PMT supply voltage and a similar range of output current.

4. Results and discussion

4.1 Demonstration

Measurements were taken in the potential core of the heated jet. The luminescence excitation waveform used is the half-sine wave, which is shown in section 4.2 to offer an improved precision over the two other waveforms. Burst signals acquired during the transits of two single particles corresponding to two different flow conditions are shown in Figs. 3(a)–3(f). The gas temperature is 293 and 840 K, and the flow velocity is 2.1 m/s and 6.2 m/s respectively. These burst signals consist of oscillating light intensities having a Gaussian envelope due to the transit of the particle through the Gaussian intensity distribution of the laser beams. The duration of the burst of the three signals is dictated by the particle transit time, which is inversely proportional to the flow velocity. This is readily observed by comparing Figs. 3(a) and 3(b) for the low velocity case and Figs. 3(d) and 3(e) for the high velocity case. The Doppler bursts occur as a result of the Mie scattered light from a particle crossing the fringes formed at the intersection of the two 514.5nm beams. Its modulation frequency, which is the Doppler frequency, is dictated by the fringe spacing and the particle velocity. As shown in Figs. 3(a) and 3(d), the Doppler frequency increases with the velocity. The 375 nm Mie scattering burst is modulated at a fixed frequency of 1 MHz set by the excitation laser waveform. As shown in Figs. 3(c) and 3(f), the resulting luminescence signal oscillates at the same frequency but is delayed with respect to the 375 nm Mie scattering burst due to the lifetime of BAM:Eu^{2+} at that particular flow temperature. As shown in Figs. 3(b) and 3(e), fewer cycles are observed at higher flow velocities. The number of cycles, N_c, of modulated incident light scattered by a particle crossing the UV beam is proportional to the particle transit time, $t_{transit}$, ($N_c \sim t_{transit} f_M$). With increasing flow temperatures a phase shift and an increase in the contrast of the luminescence burst in terms of modulation depth is observed due to decrease in emission lifetimes relatively to the fixed period of the modulated excitation. This is readily visualised in Figs. 3(c) and 3(f), where the luminescence and 375 nm Mie scattering bursts at the two conditions are plotted within a shorter time window.

Fig. 4 shows the measured phase shift of BAM:Eu^{2+} as a function of flow temperature. The error bar represents 1 single-particle standard deviation from the evaluation of phase shifts for ~ 100 bursts produced from individual particles crossing the probe volume in the constant temperature jet core. Results indicate that the phase shift is insensitive to changes in temperature below 600 K for this specific phosphor. A strong temperature dependence above 600 K is shown which is also in agreement with previous measurements of the temperature-dependence of the BAM:Eu^{2+} emission lifetime in the time domain [13].
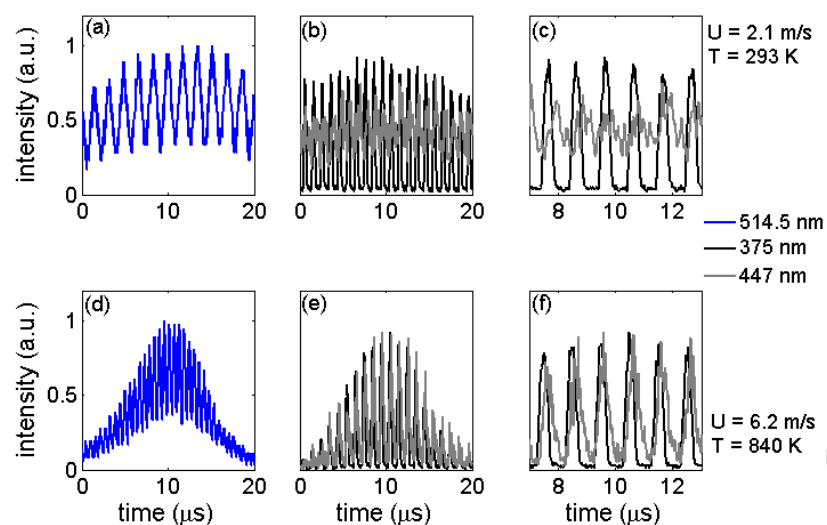
Fig. 3. Signals from a BAM:Eu^{2+} particle normalised to the maximum intensity based on using half sine-wave (HSW) excitation modulation waveform at frequency 1MHz: At flow condition 293 K, 2.1 m/s; (a) 20 μs time window of Doppler burst (514.5 nm), (b) 20 μs time window of phase-shifted luminescence (447nm) burst relative to the Mie scattering (375nm) burst ,and (c) 6 μs time window of phase-shifted luminescence (447nm) burst relative to the Mie scattering (375nm) burst. At flow condition 840 K, 6.2 m/s; (d) 20 μs time window of Doppler burst (514.5 nm), (e) 20 μs time window of phase-shifted luminescence (447nm) burst relative to the Mie scattering (375nm) burst, and (f) 6 μs time window of phase-shifted luminescence (447nm) burst relative to the Mie scattering (375nm) burst. The wavelengths are written in terms of the central wavelengths of the spectral filter used. A moving average filter of 4 sample points is applied to the 447nm signals in (b) and (c) for better visualisation.
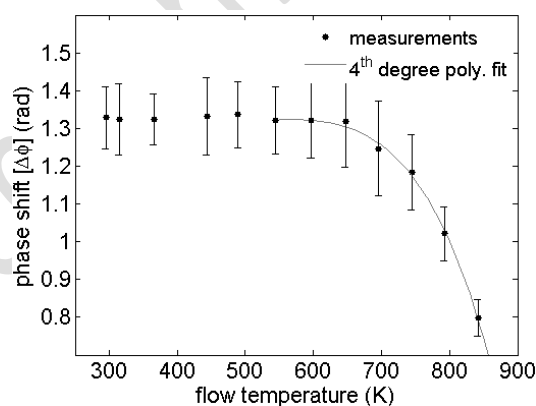

Fig. 4. Phase shift as a function of flow temperature. The error bars represent the standard deviation of 100 measurements of single particles.

Calibration data was obtained from a fourth order polynomial fit through the data points, as shown in Fig. 4, and then used to obtain temperatures from the measured phase shift data. The large standard deviation in the measured phase-shift at temperatures below 650 K, where the luminescence decay time remains constant (~ 1 μs) is expected to be largely caused by sampling noise, visible on Fig. 3. A high supply voltage is applied to PMT3 in order to detect the weak luminescence signals from individual phosphor particles. It is also possible that particle-to-particle variations in the luminescence properties cause part of the observed fluctuations.

However, to the authors' knowledge, no study on the influence of size and/or shape on the luminescence lifetime of BAM:Eu^{2+} have yet been reported.

Over the range 696 -841 K, the mean deviation or residual of the estimated mean flow temperatures to the fit is 3.8 K (0.53 %), with a maximum deviation of 9.6 K. At 795 K, the single-particle temperature precision is 19 K or 2.4%, which is more than a 3-fold improvement in comparison to measurements taken at 624 K with the intensity ratio approach described in [6]. At 841 K, the highest temperature reached by the flow system, the precision is as high as 8.6 K (1.0 %).

4.2 Choice of excitation waveform

For a particular luminescence response, a sinusoidal excitation may not be optimal and other waveforms (square wave, half sine wave) may offer a better performance [15]. The phase shift measurements were therefore repeated for two other excitation waveforms, shown in Fig. 1, and the results in terms of measured phase shift and precision are shown on Table 1. While the average phase shift seems relatively insensitive to the excitation waveform, it can be seen that the precision offered for the half sine excitation waveform is significantly better than for the full sine wave, and slightly better than for the square wave.

Table 1. Average and standard deviation of the measured phase shift for three excitation waveform, square wave (SqW), half sine wave (HSW) and full sine wave (FSW). The second row for each flow temperature corresponds to a repeated measurement set.

Mean flow Temperature	Mean Phase Shift			Standard Deviation		
	SqW	HSW	FSW	SqW	HSW	FSW
291 K	1.3042	1.3373	1.3368	0.1376	0.1012	0.1884
	1.3211	1.3391	1.3120	0.1192	0.1213	0.1659
815 K	0.9830	0.9975	0.9434	0.0768	0.0737	0.1276
	0.9685	0.9905	0.9578	0.0794	0.0790	0.1108

Furthermore, it should be noted that a key advantage of evaluating the lifetime in the frequency domain is that, for a phosphor with a given rate of change of the lifetime with temperature, the sensitivity of the phase shift to temperature can be adjusted with the choice of modulation frequency by virtue of the inverse tangent (tan^{-1}) function. To illustrate, Eq. (1) can be inverted and differentiated as in:

$$\frac{1}{\Delta\Phi}\frac{d\Delta\Phi}{dT} = \frac{1}{\tan^{-1}(2\pi f_M \tau)}\frac{2\pi f_M \tau}{1+(2\pi f_M \tau)^2}\times\frac{1}{\tau}\frac{d\tau}{dT} \qquad (3)$$

Eq. (3) shows that by decreasing the modulation frequency, the first two terms on the right hand side are a non-increasing function for $2\pi f_M \tau$ (which is always positive), resulting in an increase of the sensitivity of the phase shift for a fixed sensitivity of the lifetime. For a fixed transit time of the particles, the decrease in the modulation frequency reduces the number of cycles for the phase evaluation, but the number of samples per burst remains constant. For the BAM:Eu^{2+} particles used here, it may be estimated that reducing the modulation frequency to 250 kHz will increase the sensitivity of the phase-shift at 800 K by a factor of 4. For the flow

conditions of Fig. 3 (6.2 m/s), the number of cycles would be reduced from 20 to 5. Subject to the accuracy of the phase shift determination relative to the number of recorded cycles, there is room for optimising the modulation frequency in a given measurement situation.

4.3 Evaluation of potential interference from in-phase signals

To get a closer look at the relationship between phase shift and lifetime, the powder was excited with 100 kHz square wave excitation (5 µs on, 5 µs off), which allows the measurement of both the phase shift in the frequency domain, and the luminescence lifetime in the time domain (during laser off) from a single luminescence waveform. This would rule out any influence of differing pulse duration, and/or average excitation power. For these measurements, a PMT supply voltage of 224 V was used, resulting in a lower gain that for the gas-phase measurements. The measured phase shift is 0.5801 rad, while the phase shift derived from the lifetime measured during 0.1 – 4µs (1.16 µs) from Eq. (1) is 0.6298 rad. At this stage, it is not possible to determine whether the remaining difference (~ 8 %) is due a multi-exponential nature of the luminescence transition or to remaining in-phase signals.

Therefore, the mono-exponential character of the luminescence transition was assessed by comparing the lifetime derived from a 0.1-0.5 µs time window, to that derived from a 0.1-4 µs time window using the excitation waveform described above. Here t = 0 s refers to the falling edge of the excitation pulse (fall time <100 ns). The 0.1 -0.5 µs lifetime is 1.1 µs, which is 5 % shorter than the 1.16 µs lifetime mentioned above, which hints at a weak multi-exponential character of the decay waveform, and which may account for the difference between the frequency domain phase shift, and estimated phase shift derived from time domain lifetime measurements. In [16], multi-exponential decay fitting of the luminescence lifetime at different emission wavelengths gave evidence of a faster decay component with a lifetime of ~270 ns. In the frequency domain, for multi-exponential luminescence decays, the estimated lifetime from the measured phase shift using Eq. (1) decreases with increasing modulation frequency [11,17]. By varying the modulation frequency, measuring the phase shift and converting it in lifetime using Eq. (1), the same tendency was also observed, as shown in Fig. 5 which further suggests a multi-exponential decay nature of the luminescence of BAM:Eu^{2+}.

These investigations demonstrate that the phase shift measured from individual phosphor particles and used to derive the temperature is very closely related to the decay time of the 4f5d luminescence transition in BAM:Eu^{2+}, and that interferences from in-phase signals are in this case negligible.
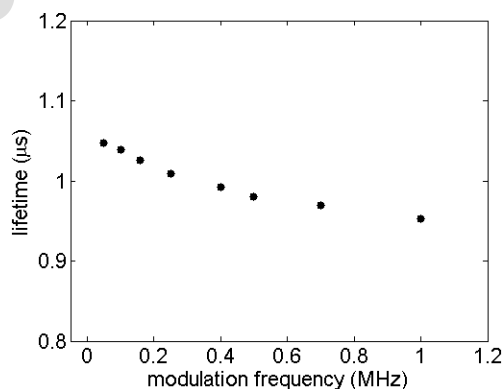

Fig. 5. Dependence of luminescence lifetime on modulation frequency using Eq. (1) and phase shift data for the HSW.

The state of assemblage of the phosphor particles, whether they are dispersed in the fluid or collected as a stack of powder, may have an influence on the luminescence characteristics. A comparison of the phase shift measured in the powder, and in the gas phase, where the number density is comparatively very low, was performed for the same detector operating parameters. When the voltage of PMT 3 is set to 570 V, the value used in the gas phase measurement, the measured phase shift for a 1 MHz modulation is 1.32 rad which is in excellent agreement with the phase shift measured in the gas-phase at room temperature (Table 1). This result supports that the conclusions above on a negligible impact of in-phase signals also apply to the fluid measurements. It also suggests that a calibration in the powder may be suitable to calibrate gas-phase measurements. This is in contrast with ratio-based measurements, which often require in-situ calibration, as discussed in an upcoming review article [18]. In this regard, measurements at 1 MHz using the stack of powder were performed with increasing the PMT gain (set by the supply voltage) from 182 V to 570 V. It was noted that the phase shift decreases with increasing gain from 1.45 to 1.32 rad, so it is necessary to operate on the same supply voltage during experiments.

5. Conclusions

A new measurement concept for gas-phase temperature measurements based on the phase-shift of the luminescence emission under modulated excitation was developed to accomplish measurements of flow temperatures using $BAM:Eu^{2+}$ phosphor particles. The choice of $BAM:Eu^{2+}$ for this proof of concept study is motivated by the fact it is a well characterised thermographic phosphor with only a weak multi-exponential decay character, a high quantum yield and that it is directly excitable by an inexpensive (<10 k€) 375 nm diode laser. By seeding $BAM:Eu^{2+}$ particles in an electrically heated air jet, this approach is shown to provide high-precision temperature measurements at elevated temperatures beyond 700 K where the lifetime of $BAM:Eu^{2+}$ is sensitive, i.e. 8.6 K at 841 K. For high temperature applications, this is a significant improvement compared to the intensity ratio measurements where the temperature sensitivity decreases with increasing temperatures. Using various excitation schemes, it was shown that the phase shift is independent of the shape of the excitation waveform, and this quantity is closely related to the luminescence lifetime of $BAM:Eu^{2+}$, with little influence from interference from in-phase signals such as scattering and fast luminescence. In addition, the comparison between measurements in the powder and in the fluid suggests that this phase-shift approach may not require an in-situ calibration, likely a significant advantage in many practical applications. Another promising feature of this approach is the possibility to tune the sensitivity of the phase shift to temperature via the modulation frequency.

It is worth noting that there is another method by which flow measurements using the phase shift evaluation method could be made that does not require a temporal intensity modulation of the laser light to generate the oscillatory excitation waveform needed to measure the phase shift. By forming an LDV probe volume using two intersecting continuous wave laser beams of sufficient coherence length (much greater than the coherence length of the diode laser used in this work (~ 100 μm)), each particle traversing the probe volume would experience modulated excitation due the fringes present in the probe volume. As before, comparison of the Mie scattered and luminescent emission would reveal a phase shift related to temperature whilst the fringe crossing frequency would provide the velocity. By this approach, temperature and velocity could be obtained from a single LDV probe volume and with a single laser, albeit an UV one. Despite the elegance of this approach, there are some complications. The method provides little control over the fringe spacing and hence the excitation frequency for the phosphor. Moreover, though the excitation frequency (Doppler frequency) would always be known from the Mie scattered signal, this frequency might be different for each particle due to velocity fluctuations. Although, in this paper, we concentrated on the simpler and more controllable approach of modulating the excitation laser beam, so that the modulation frequency

is independent of the particle velocity, the above mentioned alternative certainly merits further investigation.

The phase-shift based temperature measurement concept demonstrated in this paper is promising. The BAM:Eu^{2+} phosphor shows high-sensitivity in the range 700-850 K which is of interest for in-cylinder studies of internal combustion engines [2], with the additional benefit that the phase-shift is in principle insensitive to pressure and oxygen concentration. Also, phosphors having a lifetime varying over a lower temperature range (300-700 K) will be better suited to turbulent heat transfer investigations and should be explored. An alternative could be another Eu^{2+} doped phosphor with an onset of thermal quenching at lower temperature. The lifetime of $(Sr,Mg)_2SiO_4:Eu^{2+}$, for example, starts decreasing at 450 K[13]. The phosphor $(Ba,Sr)_2SiO_4:Eu$, developed for white LED is shown to start quenching at room temperature and is therefore promising as well [19].