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RAPID COMMUNICATION

Investigation of potential laser-induced heating effects when using thermographic phosphors for gas-phase thermometry

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Abstract The spectral emission from thermographic phosphors in free flow and its dependence of laser energy per cross section area (laser fluence [$\mathrm{J/cm^2}$]) has been investigated. Temperature measurements in gaseous flows using thermographic phosphors require higher laser energy than measurements performed on surfaces, due to lower particle density. A troublesome systematic error associated with high fluences would be introduced if the excitation laser heats the particles. In the presented work, three different types of the thermographic phosphor $\mathrm{BaMg_2Al_{10}O_{17}}$:Eu (BAM) are investigated. Spectra of the phosphorescence are achieved for a range of laser fluences. The results show no indications of the laser heating the particles, making further development of phosphor thermography in free-flow applications feasible.

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1 Introduction

Thermographic phosphors (TP) are ceramic crystal substances, often in the form of a fine powder. Usually they are doped with some kind of rear earth metal, which gives them specific phosphorescence properties. These phosphorescence properties are sometimes temperature dependent, which makes TP useable in temperature measurement applications. The use of TP is well established for measuring temperature on surfaces [1–5]. By applying laser-induced phosphorescence (LIP), temperature can be monitored by registering the change in phosphorescence lifetime or the change in spectral profile. Both point- and two-dimensional measurements are possible. The point measurement approach usually utilizes the temperature dependence of the phosphorescence signal lifetime to retrieve temperature information [6]. By using a high-speed framing camera it is also possible to do 2D measurements using this property [7]. Exploration of the temperature dependence of the spectral profile makes it possible, with the use of suitable optical filters, to do 2D temperature measurements on a surface either through the use of two separate detectors or a single detector equipped with an image doubling device (stereoscope) [8]. With a suitable choice of phosphor material, measurements can be carried out at temperatures from cryogenic to 2000 K. Due to interference with background Planck radiation at higher temperature, phosphors with emission in the blue

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spectral region are preferable in order to increase the signal-to-noise ratio. In addition, short lifetime of the phosphorescence gains the possibility to time gate for background discrimination [9].

Lately, attempts have been made to seed TP in gaseous flows in order to investigate the feasibility of temperature measurements in gas volumes [10]. Having access to such a technique with its 2D capability is highly desirable, e.g., in combustion engine research. In gas-phase LIP measurements a laser beam is preferably formed into a laser sheet in order to establish spatial resolution in the depth dimension. A higher laser fluence compared to what is applied for surface thermometry is necessary in order to achieve a sufficient signal from the relatively diluted particle flow. In a typical engine application, a laser sheet could be about 20-mm high and 0.5-mm thick. This limits the laser pulse energy to a maximum in the region of 100 mJ due to the risk of damaging optical components. These numbers result in an upper limit for the fluence of 1.3 $\mathrm{J/cm^2}$ in such practical applications. When reaching high laser fluences, an obvious, but still highly relevant, question is whether the laser itself is heating the particles or not. For example, laser-induced incandescence, LII, a standard technique for soot diagnostics, is based on laser heating of particles. If such heating should occur also for the phosphor particles, this would make measurements in gaseous media virtually impossible. To investigate this, an air flow at room temperature seeded with the thermographic phosphor BAM is illuminated with laser radiation, and the phosphorescence signal is recorded with a spectrometer at a range of different laser fluences.

$\mathrm{BaMg_2Al_{10}O_{17}}$:Eu, or BAM, emits very strong phosphorescence in the blue spectral region when illuminated with laser radiation of 355 nm. This emission originates from the $4f^65d \rightarrow 4f^7$ transition of $\mathrm{Eu^{2+}}$ [11] and has its peak at about 450 nm, which broadens towards shorter wavelengths with increasing temperature, see Fig. 1 [12]. The ratio of the phosphorescence intensities, collected through a 40-nm wide (fwhm) interference filter centered at 400 nm and a 10-nm wide (fwhm) interference filter centered at 456 nm, will show a temperature dependence as shown in Fig. 2. Using these two filters, BAM is very suitable for achieving 2D temperature images of a coated surface. Because of its high signal intensity, BAM is also beneficial in free-flow applications. It is also favorable to use BAM in this kind of investigation, since it is sensitive at room temperature. The short lifetime of the phosphorescence, about 2 µs at room temperature and about 10 ns at 1100 K, also make BAM suitable for measurements with high temporal resolution, whether the measurements are done on surfaces, in free flow, or in sprays [10, 13]. With such a short lifetime it is possible to resolve sub-millimeter structures at velocities up to 300 m/s.

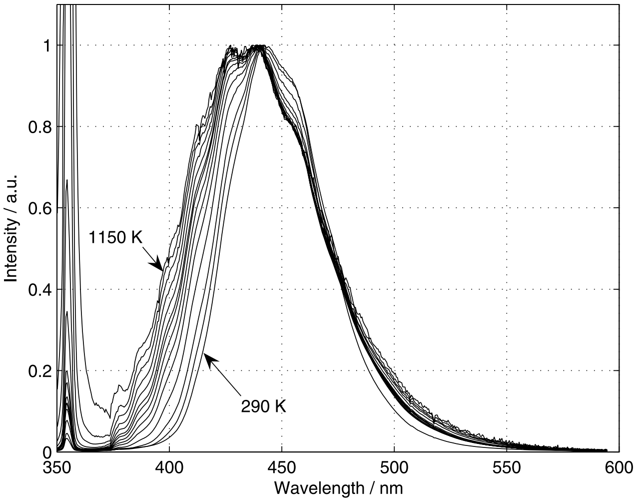

Fig. 1 BAM emission spectrum at different temperatures when excited with 355-nm laser radiation. The exposure time of the ICCD camera connected to the spectrometer was 8 µs

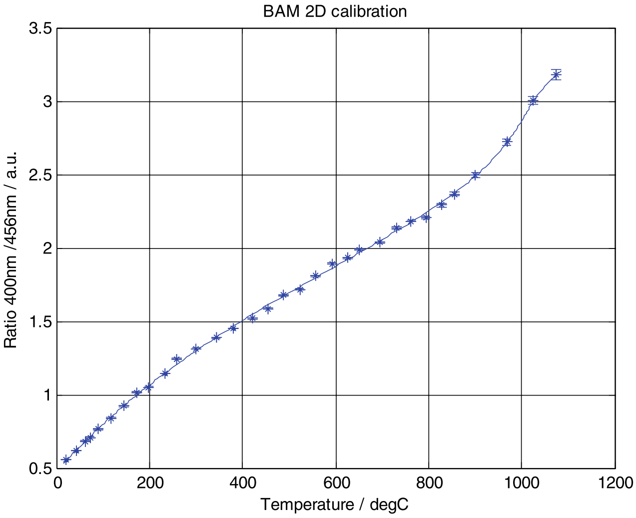

Fig. 2 Calibration of BAM for 2D measurements. The ratio of the intensities collected through the 400 ± 20-nm interference filters to that collected through the 456 ± 5 nm interference filters shows a significant temperature dependence. The filters were placed on a stereoscope connected to an ICCD camera with the exposure time of 8 µs

A criterion for LIP temperature measurements using TP is optical access, both for the exciting laser radiation and for the detection of the phosphorescence signal. However, using TP as a fine powder seeded in gaseous flow, in such an environment optical access might become troublesome due to particles sticking onto surfaces. One way to reduce this issue is to use coated phosphor particles. From the manufacturer (Phosphor Technology) it was possible to order two types of “anti-stick” coating for BAM, $\mathrm{SiO_2}$ coating, and $\mathrm{Al_2O_3}$ coating. Experiments where these different types of BAM were applied on surfaces show that the phosphorescence spectrum is not affected by the coating. From experience BAM coated with $\mathrm{SiO_2}$ seems to stick the least. In the free-flow experiments, all three types are investigated for possible laser-induced heating.

The mean size of the particles used in the experiments is 4.2 µm and, the density about 3.7 $\mathrm{kg/dm^3}$ according to the supplier. The small size allows the particles to follow possibly turbulent flow well when the Stokes number is much smaller than 1.0, which is the case in most engine applications and under the present experiment conditions. The coating with SiO_2 and Al_2O_3 is of nanometer-scale thickness and has a negligible effect on the particle size.

Another criterion for using seeded particles in a gaseous flow to measure its temperature is that the particles establish local thermal equilibrium with the surrounding gas on a time scale comparable to the temporal temperature variations. Assuming the internal temperature distribution in the phosphor particle to be homogeneous, this relaxation time is estimated by solving the heat convection equation [14]

$$hA(T_\infty - T) = c_p \rho_p V \frac{\mathrm{d}T}{\mathrm{d}t},$$

where h is the convection heat transfer coefficient, A is the surface area of the particle, T_∞ is the gaseous temperature, T is the particle temperature, c_p is the specific heat of the particle material, ρ_p is the density of the particle, and V is the particle volume. With the initial condition that T_0 is the initial particle temperature at $t = 0$, the solution is described by

$$\frac{(T - T_0)}{(T_\infty - T_0)} = 1 - e^{-\frac{t}{\tau}},$$

where τ is the so-called time constant,

$$\tau = \frac{\rho_p c_p V}{hA}.$$

The time by which the phosphor particles establish local thermal equilibrium in gaseous flows with rapidly varying temperatures depends very much on the current conditions, such as temperature of the surrounding gas, particle size, particle substance, turbulence, and, in particular, air properties such as density, heat capacity, viscosity, and thermal conductivity, which all are temperature dependent. The heat transfer coefficient h can be calculated using the following relation [15]:

$$\mathrm{Nu} = \frac{hd}{k} = 2 + 0,6 \cdot \mathrm{Pr}^{1/3} \cdot \mathrm{Re}^{1/2},$$

where Nu is the Nusselt number, d the particle diameter, k the thermal conductivity of the gas, Pr the Prandtl number, and Re the Reynolds number.

For a particle initially at room temperature, this would result in a relaxation time of 170 µs for the particle to reach 90% of a surrounding air temperature of 700 K. However, the model is based on spherical particles, which have the smallest possible surface area to volume relation, thus the relaxation time is thought to be shorter for real phosphor particles. Also the assumption that the particles have an even internal temperature distribution is simplified. Initially there most probable is a temperature gradient over the particle. The collected signal is biased towards phosphorescence originating from the particle surface, where the temperature more closely corresponds to the gas temperature.

The argumentation above should be considered as a rough estimation. Future work calls for both theoretical and experimental investigations regarding phosphor particles establishing thermal equilibrium and the timescales involved.

2 Experimental

In the experiments a 10 Hz pulsed Nd:YAG laser operating at its third harmonic producing 355-nm radiation was used as excitation source. The exiting beam profile was circular (Gaussian) with a diameter of about 8 mm. A single cylindrical lens ($f = 1000$ mm) was used to focus the beam in the vertical plane. 500 mm after the lens, the narrowed beam crossed an air flow that was kept at room temperature and seeded with phosphor particles. Thus, at this position, the laser beam had the shape of an ellipsoid with the size of 8 mm in the vertical direction and 4 mm in the horizontal direction. With the laser pulse energy set at 400 mJ, a corresponding fluence of 1.5 $\mathrm{J/cm^2}$ was produced. The phosphorescence signal, collected by a lens system into a spectrometer, was detected with an ICCD camera. Spectra were recorded for a range of fluences from 0.1 to 1.5 $\mathrm{J/cm^2}$.

3 Results and discussion

Figure 3 shows emission spectra of uncoated BAM at three different laser fluences. As can be seen, the spectra are completely overlapping, showing no indication of temperature change. The emission spectra of BAM with SiO_2 coating and with Al_2O_3 coating, showed the exact same behavior as that of uncoated BAM at different fluences and are therefore not included in the paper.

In a real measurement situation, the spectrum is not recorded, but the area of interest is being photographed through two adequate filters. In order to simulate what the ratio of the intensities through the two filters would be, the recorded spectra were multiplied by normalized transmission spectra for the two filters mentioned above. The resulting ratios as functions of laser fluence are shown in Fig. 4. As can be seen, there is no tendency of the ratio changing with laser fluence and therefore neither any induced temperature change.

Using excessively high laser fluence (above 10 $\mathrm{J/cm^2}$), a completely different spectrum is detected, as compared to

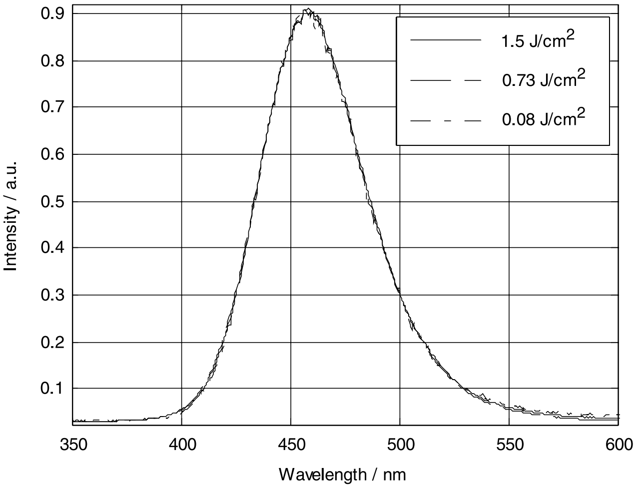

Fig. 3 BAM emission spectrum at three different laser fluences

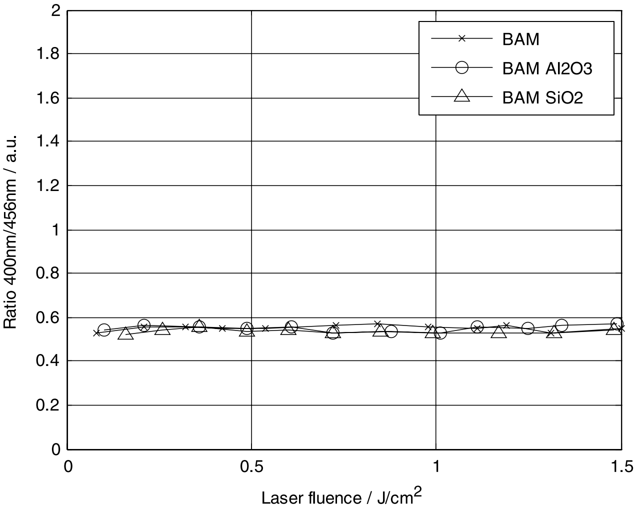

Fig. 4 Evaluated ratio with respect to the transmission of the 400 ± 20-nm and 456 ± 5-nm interference filters, as a function of laser fluence for all three types of BAM

the phosphorescence spectrum, with discrete peaks more resembling a laser-induced breakdown spectrum, LIBS. The spectra from the three different BAM types differ slightly. The SiO_2-coated BAM seems to be a bit more resistant to higher laser fluence, since the intensities from the distinct peaks are relatively lower than the ordinary phosphorescence signal, compared to the other BAM types. BAM coated with SiO_2 is thus preferable, not only because of its better “anti-stick” properties, but also because it is more resistant to higher fluences.

4 Conclusion

The experimental results show that high laser fluence, up to $1.5\ \mathrm{J/cm^2}$, does not tend to heat phosphor particles when used in gaseous flow measurements. In this kind of measurements higher laser energy (compared to surface thermometry), and thus higher laser fluence, is needed to achieve a sufficient signal level due to the low particle density. Therefore heating the particles with the excitation laser is the biggest concern. In the range of typical laser fluences used in LIP applications there are no indications of the laser heating the particles of the thermographic phosphor BAM, when seeded in a gaseous flow of room temperature. This makes further development of free-flow measurement techniques promising.

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References

1. S.W. Allison, G.T. Gillies, Rev. Sci. Instrum. **68**, 2615 (1997)
2. A. Khalid, K. Kontis, Sensors **8**, 5673 (2008)
3. H. Seyfried, M. Richter, K.-H. Nilsson, M. Aldén, H. Schmidt, Collection of Technical Papers—45th AIAA Aerospace Sciences Meeting, vol. 18 (2007), p. 12794
4. T. Husberg, S. Girja, I. Denbratt, A. Omrane, M. Aldén, J. Engström, SAE 2005-01-1646 (2005)
5. J. Brubach, J.P. Feist, A. Dreizler, Meas. Sci. Technol. **19**(2), 025602 (2008)
6. L.J. Dowell, G.T. Gillies, Rev. Sci. Instrum. **62**, 242 (1991)
7. A. Omrane, F. Ossler, M. Aldén, Proc. Combus. Inst. **29**, 2653 (2003)
8. H. Seyfried, G. Särner, A. Omrane, M. Richter, M. Aldén, ASME, GT2005-69058 (2005)
9. S. Gustaf, R. Mattias, A. Marcus, Meas. Sci. Technol. **19**, 125304 (2008)
10. A. Omrane, P. Petersson, M. Alden, M.A. Linne, Appl. Phys. B—Lasers Opt. **92**, 99 (2008)
11. S. Shionoya, H. Yamamoto, W.M. Yen, *Phosphor Handbook*, 2nd edn. (CRC Press/Taylor and Francis, Boca Raton, 2007)
12. G. Särner, Doctoral Thesis, Lund University (2008)
13. J. Brubach, A. Patt, A. Dreizler, Appl. Phys. B—Lasers Opt. **83**, 499 (2006)
14. J.P. Holman, *Heat Transfer*, 8th edn. (McGraw-Hill, New York, 1997)
15. W.E. Ranz, W.R. Marshall, Chem. Eng. Prog. **48**, 141 (1952)