Breakdown of the large-scale circulation in $\gamma=1/2$ rotating Rayleigh-Bénard flow

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Discharges for lighting

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Discharges for lighting

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Abstract

The most common man-made discharge is a lamp. Even though lamps are often considered a mature technology, the discharge physics is often poorly understood. Two recent initiatives discussed here show that plasma research can help to make significant improvements. First we discuss color separation in metal halide lamps, which is a problem that prevents these highly efficient lamps from being used in more applications. Secondly a novel lamp concept is presented that may replace the current mercury based fluorescent lamps.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Starting with the wood fires in the Stone Age people make light in the dark. There has been a consistent improvement in technology. Oil lamps, candles and gas lighting have been used for centuries and since the invention of incandescent lamps our main light sources have been electrically powered. The impact of light on our lifestyle is enormous. It enables us to live and work inside buildings and to extend our active daily routine far beyond daylight hours. We use it for entertainment (television, neon advertisements), for information (computer and mobile phone screens), for safety (car and streetlights), for crops in greenhouses or simply as decoration in gardens or on buildings. However, lighting our world comes at a high environmental price. In the first place we disturb the natural cycle of plant and animal life, secondly lamps often contain environmentally unfriendly materials that should be recycled at the end of the life cycle and finally light production also produces a large amount of carbon dioxide, the main greenhouse gas. Some numbers to illustrate this [1]: worldwide about 20% of all electricity is used for lighting. The resulting CO₂ emission amounts to about 70% of the emission by passenger cars or to the total emissions of France, Germany, Italy and UK combined. These numbers show that the introduction of energy efficient lighting can make a very large impact on CO₂ emission and thus on global warming. There is large variation in lamp efficiencies as can be deduced from table 1. The well-known and commonly used incandescent lamp has about the worst possible efficiency, which explains why there is a strong push to ban it in favor of compact fluorescent lamps (also known as energy saving lamps). Note that the latter still

Table 1. Lamp efficacy and power consumption in the European Union.

Lamp type	Use	Luminous flux (lm/lamp)	Typical efficacy (lm W ⁻¹)	Light output (% of all light)	Light power consumption (%)
Incandescent	Domestic	90–3000	12	4	26
Halogen	Domestic	60–44 000	15	1	5
Compact fluorescent	Domestic	100–12 000	70	4	2
Fluorescent	Indoor, office	1000–7000	80	51	41
Mercury vapour	Street lighting	1600–58 000	50	Currently being replaced by HPS and MH lamps	
High pressure Na (HPS)	Outdoor	3500–130 000	100	34	23
Low pressure Na	Street and tunnel lighting	2000–40 000	200	34	23
Metal halide (MH)	Outdoor and factory	3300–320 000	100	34	23
LED	Specialty uses	0.01–10	60	<1%	<1%

perform worse than fluorescent tubes. It is obvious that other issues, apart from efficiency are important. The low pressure sodium lamps (the deep orange street lamps found in some countries), for example, are very efficient, but their monochromatic light renders everything grayish, so it can only be used as street lighting. Table 1 shows that nearly all lamps are based on plasma technology. The exceptions are the highly inefficient incandescent (halogen) lamps and the technologically immature light emitting diodes (LED). The latter will in the near future prove to be a very good alternative in some cases, especially as a replacement for incandescent lamps. However, even if LED technology is advancing as rapidly as predicted, electrical discharges will have to supply the major share of man-made photons for at least another two decades [2,3].

Generally two groups of discharge lamps are distinguished: low and high pressure lamps. Apart from their pressure, their principle of operation is different. A low pressure lamp is a non-equilibrium discharge, in many cases similar to the classical positive column. Light is generated by the emission of excited states, populated by electron impact excitation. Since metals have resonant states emitting in the visible they seem ideal. However in order to avoid heat losses, the gas should stay cold so most metals have insufficient vapor pressure. Mercury is the metal of choice since it has a high vapor pressure and can be easily excited. It generates light in the UV (254 and 185 nm), which can be converted into white visible light by phosphors: the tubular fluorescent tube and its cousin the compact fluorescent lamp. Mercury is a highly efficient radiator, but half the photon energy is lost in the phosphor where a UV photon is converted into a visible one. Sodium has a slightly lower vapor pressure, but its resonant line at 590 nm coincides with the maximum sensitivity of our eye. Therefore no phosphors are needed yielding a very efficient lamp, which unfortunately is monochromatic and thus of limited use.

High pressure lamps are based on arc emission. These discharges are close to local thermodynamic equilibrium and their emission lines are close to the Planck limit. Again mercury and sodium are commonly used, but since the end of the last century metal halide salts such as NaI, DyI₃ and HoI are often added to increase efficiency and color properties. High pressure lamps produce large amounts of light and do so very efficiently (see table 1). Consequently they are commonly used in city and industrial lighting.

It is clear that there is a large variety of lamps and lamp types. Also there is more than an order of magnitude in variation in lamp efficiency between the incandescent lamp and the

low pressure sodium lamp. As mentioned above, light generation comes at a high cost, so any improvement can have a significant impact. Currently, technological improvements are steadily made, but real advancement can only come from either new technologies or a significantly better understanding of the existing lamps. Examples of both cases will be discussed below.

2. Color separation in metal halide lamps

Looking at table 1 the first question that comes to mind is: why do not we always use metal halide lamps as they are highly efficient and produce white light. The reason is that the lamp has several issues. For example, it can only be operated in a specified position, e.g. vertically. If operated wrongly it becomes unstable which is seen as a disturbing flickering. It needs to cool down—the mercury vapor needs to condensate—before it can be re-ignited: not useful in the case of power failure or the accidental flick of a light switch. And it may have a non-homogeneous color: not useful for shop or domestic lighting where excellent color rendering is required. These issues are caused by discharge physics, which is only poorly understood. In past years our group has focused on understanding the color separation in discharge lamps [4–14].

In order to qualitatively describe the phenomenon let us consider the lamp in slightly more detail. A metal halide lamp is derived from the better known mercury high pressure arc discharge. An electric arc is maintained between two electrodes, which are sealed into the confining space of the lamp burner. In normal operation, the main gas in the lamp is mercury. Depending on lamp type, size and application, the pressure varies from a few to several hundred bars. The former are large lamps used as street or stadium lights, the latter are tiny lamps used in video and beamer projection systems. The temperature in the center of the arc is about 5000 K, while the wall is maintained around 1200 K. As the lamps are in the millimeter to centimeter size range, this implies that there are strong temperature gradients. A high voltage peak starts the lamp in a noble gas, but after a few seconds the mercury starts evaporating. Due to its lower ionization energy, the arc soon becomes a mercury discharge. As mentioned before mercury discharges are very efficient but the energy of mercury emission is only partly utilized. Metal halide lamps use the efficiency of a mercury discharge, while directly emitting visible photons. This is achieved by adding a mixture of metal halide salts (such as DyI_3 , ScI_3 , CsI_2 or NaI) to a mercury arc discharge. Obviously, in the hot central arc region, the salt will dissociate and the metals will be excited and ionized. These metals are extremely efficient radiators. Even though the metal additive density is much less than the mercury density, most of the light is emitted by the metals in the visible region. As a result, metal halide lamps have better power efficiencies than most other lamp types.

A metal halide lamp with color separation has different colors along its axis. Figure 1 shows an example of color separation in a lamp containing DyI_3 . Whereas the bottom of the lamp is white with a reddish glow around it, the top is greenish. The cause is a complex interplay of diffusion and convection resulting in a non-homogeneous distribution of the radiating metals. Let us concentrate first on the radial direction. Going from the wall at about 1200 K to the arc core at 5000 K metal halide molecules will first dissociate and then ionize. The resultant density gradients cause strong diffusion fluxes, which are counteracted by the back-diffusion of other species: near the wall molecules diffuse inwards and atoms outwards, while near the center atoms move inwards as the ions diffuse out. The diffusion speed of the molecules is significantly smaller than that of the atoms due to their size and mass difference. Ions and atoms have similar masses, but ions diffuse faster due to ambipolar diffusion. In a stationary state with equal in- and outward fluxes, a lower speed is balanced by a higher partial pressure. The

Figure 1. (Left) Picture of a DyI₃ lamp with color separation, visible as a red glow near the bottom. (Right) Artist impression of the Dy flux (in any chemical form) inside the lamp. The combination of axial and radial transport results in a higher Dy density near the bottom of the lamp.

total effect is called radial segregation and results in a reduction of the partial metal pressure (in any chemical form) near the lamp center.

In addition to diffusion, there is also convection in the lamp. In vertically operating lamps, convection causes an upward flow in the hot central arc region, which returns downwards along the cooler walls. The minority metal salt species follow the dominant mercury convection flow. However, due to radial segregation the metal that rises in the center diffuses outwards to the wall and is transported down. The combined effect of convection and radial segregation is a decreased metal density in the upper part of the lamp (figure 1). Such axial segregation results in a non-homogeneous emission, called color separation, shown in figure 1. The effect has a maximum if convective and diffusive flows are of equal magnitude. High convective velocities result in a good mixing of species eliminating radial segregation. In the absence of convection, the lamp emission will be axially uniform but radial segregation is maximal.

This explanation, first described by Fischer [15], yields a reasonable qualitative description of the effect. However, quantitative models, needed to increase lamp efficiency, are not available. Discharge calculations of diffusion and convection in complex reactive mixtures with high temperature gradients are difficult and many thermodynamic data are not known. In addition the electrodes, the lamp geometry and wall material also affect the lamp behavior. Experiments are necessary to provide input data and verify any model results. In order to facilitate comparisons a reference lamp has been defined [11], which is used in the work described here. This reference lamp has a 2 cm long, 8 mm diameter cylindrical quartz burner and contains apart from mercury only DyI₃ as a metal halide salt. This allows for easy experimental access as well as modeling simplicity.

To be able to unravel the complex interaction between convection and diffusion, extended experiments under zero and hyper-gravity have been performed. In the absence of gravity, convection is eliminated, so the effect of diffusion can be studied undisturbed and

Figure 2. Lateral profile of the atomic Dy line at 642.19 nm, measured by means of emission spectroscopy. The lamp contains 4.2 mg DyI_3 and 10 mg Hg. The lamp input power is 150 W. The measurement is taken near the top of the burner. The intensity near the top of the lamp is higher at 10 than at 1 g due to reduced axial segregation.

Figure 3. Line-of-sight 2D ground state atomic Dy density distribution at 1 g (left) and 10 g (right). $y = 0$ is the bottom and $y = 400$ is the top of the 20 mm high lamp; $x = -140$ and $x = 660$ are approximately the positions of the wall (inner diameter 8 mm). The Dy density values are given in m^{-2} on a logarithmic scale. Note that the scales of both graphs are different. In both cases axial segregation is observed, but at 10 g, but the distribution is more homogeneous.

compared with the model results. Parabolic free falls in an airplane have been performed in an airbus of the European Space Agency (ESA) and extended micro-gravity experiments were done in the International Space Station [4–7].

In order to enhance the dynamical range of the experiments to higher gravity values, a centrifuge setup has been built [13, 14]. The lamp, including optical emission spectroscopy and imaging laser absorption spectroscopy setups, is placed in a rotating gondola. The resultant apparent gravity can be varied continuously from 1 to 10 g. The result of the increased apparent gravity is an enhanced convection flow within the lamp, resulting in a redistribution of the radiating metal. Figure 2 shows optical emission measurements in the top of the lamp for 1 and 10 g. In figure 3 the Dy density in the lamp is mapped using absorption spectroscopy. In both cases the data show that increased convection results in less axial segregation due to better mixing. The more evenly distributed Dy inside the lamp results not only in a more homogeneous color rendering of the lamp but also in a higher overall lamp efficiency. It is currently beyond

Figure 4. Emission spectrum of InBr showing atomic lines at 410 and 450 nm as well as a molecular band between 360 and 390 nm. These emission lines are closer to the visible so the lamp is potentially more efficient than a Hg lamp.

our capabilities to fully understand and model a commercial metal halide lamp with a complex shape and chemistry [12]. Therefore, the measurements have been performed on a reference lamp with a simple geometry and chemistry. The absence of convection under micro-gravity conditions greatly simplifies the problem. The measurements reveal a great deal about lamp behavior, which needs to be reproduced by any valid lamp model. Qualitatively the lamp behavior is well understood, and the measurements supply ample data for quantitative model validation. In addition, the measurements clearly show that there is still room for efficiency gain. Obviously, changing gravity is not a practical solution, but it helps to identify how to optimize discharge parameters, so we can illuminate our world using less energy.

3. Molecular discharge lamps

Table 1 shows that approximately half of all light is produced by low pressure gas discharge lamps: the well-known fluorescent tubes in offices and schools. As mentioned before, Hg in these lamps emits at 254 and 185 nm so half the photon energy is lost by the conversion of the UV photons to visible light by the luminescent phosphors on the wall. Therefore novel Hg-free discharge chemistries are considered that emit light in the near-UV or directly in the visible region.

As is commonly known, mercury is unhealthy and thus mercury-free lamps are desirable from this point of view. However, current mercury lamps are highly efficient. In assessing the ecological impact it should be noted that there are small amounts of mercury in the average fuel mix of power plants. During the long lifetime of these discharge lamps, generally about five to ten times more mercury is released into the atmosphere by electricity plants supplying the lamp power than is present in the lamp. Moreover, the mercury in these lamps is easily recyclable. Therefore, it is of no use to introduce a mercury-free lamp if this results in a loss of lamp efficiency.

Several aspects need to be considered in choosing a discharge chemistry for light emission. Obviously efficiency is a major concern, but also chemical stability, ease in handling and discharge breakdown and operating voltages. Noble gases and gas mixtures have been studied in detail. And in some cases relatively efficient lamps can be produced [16]. Simple molecular gases such as N_2 and H_2O are also very easy to handle. Moreover they emit in the near-UV, similar to mercury. However, it seems that the efficiency that can be obtained with these molecules is limited [17]. Indium- and galliumhalides are interesting candidates as they reach significant densities at relatively low temperatures [18–20]. In this work we focus on understanding light production in InBr. The spectrum of InBr (figure 4) exhibits strong atomic

Figure 5. The time dependent 266 nm laser induced fluorescence signal of InBr observed at 364.52 nm (B–X $\Delta v = 0$ transition). With (top) and without plasma (bottom). The amplitude of the signals is scaled and shifted.

lines as well as molecular radiation that can be used in lighting. The wavelengths are close to the visible region, so conversion into white light will result in much less energy loss than using Hg emission.

The discharge is produced inside a sealed quartz cylinder (300 mm long, 38 mm diameter). The lamp is filled with argon as a buffer gas and a few mg InBr salt. In order to produce sufficient InBr vapor pressure the lamp is placed inside a oven made of ceramic quartz radiators. Thermocouples close to the lamp are used to keep the lamp at the desired temperature. A capacitively coupled 13 MHz discharge is produced using external electrodes at each end of the lamp. Hence there are no electrodes in direct contact with the discharge. The discharge is studied spectroscopically using a spectrograph equipped with a photomultiplier and a CCD camera. Yang *et al* [21] showed that a LIF signal is generated in the 350–400 nm range using the fourth harmonic of a Nd–YAG laser. We use a 10 mJ pulse⁻¹, 20 Hz, 266 nm Nd–Yag laser to excite the InBr. The integrated as well as the spectroscopically and time resolved responses are monitored.

At low temperatures the InBr vapor pressure is low and the discharge is essentially an Ar plasma. Above 450 K, the discharge changes color and effectively becomes a InBr discharge (figure 4). Using 266 nm photons ground state InBr molecules can be excited to higher excited molecular states. From these states the lower states emitting between 360 and 390 nm can be populated. The nature of this coupling is unknown, but is thought to be an important factor for the efficiency of low pressure metal halide lamps, like the InBr discussed here. For higher laser powers also two-photon excitation occurs. This results in photodissociation of InBr and production of excited In. This can be observed as resonant emission at 410 and 451 nm. The molecular LIF signals are relatively strong, which indicates that there is a very good coupling between the higher states excited by the 266 nm laser photons and the lower excited A and B states whose radiation is observed. The observed spectrum is in reasonable agreement with the data reported by Yang *et al* [21]. The general peak-structure is similar to the emission spectrum shown in figure 4. But both the relative magnitude of the various peaks as well as their shape show significant deviations. Further research is needed to study these differences and pinpoint the coupling mechanisms between the various levels. Figure 5 shows the time dependent behavior of the LIF signal at 364.5 nm (0–0 B–X transition) in gas vapor and in the

presence of a plasma. It is clear that both the rise time as the decay time of the molecular response is much slower than the laser pulse length. Furthermore, it is interesting to note that there is a large difference in the temporal behavior when the plasma is switched on. In the presence of a plasma the LIF signal shows both a longer rise as well as a longer decay time. A careful analysis of these signals will give insight into the molecular processes coupling the various excited states, which in turn can explain the high emission efficiency of InBr discharge lamps. In the future, this might result in a novel Hg free lamp.

4. Conclusions

Electrical discharges are the main source of artificial light. To increase their efficiency and reduce the ecological impact of light generation it is necessary to improve existing light sources as well as to develop novel ones. Many of the complex processes inside a lamp are still poorly understood and a better understanding can help to get better lamps. Two examples, color separation in metal halide lamps and molecular low pressure discharge lamps, are presented to show how plasma research can help improve lamp development.

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References

- [1] 2006 *Light's Labour's Cost* (International Energy Agency, Organisation for Economic Co-operation and Development) ISBN 92-64-10951-X
- [2] 2006 *Solid-State Lighting Research and Development Portfolio* (Navigant Consulting) <http://www.netl.doe.gov/ssl/PDFs/SSLMultiYearPlan.pdf>
- [3] Krames M 2007 Progress in High power light-emitting diodes for solid state lighting *Proc. 11th Int. Symp on the Science and Technology of Light Sources (Shanghai, China)* ed M Q Liu and R Devonshire p 571
- [4] Flikweert A J, Nimalasuriya T, Groothuis C H J M, Kroesen G M W and Stoffels W W 2005 *J. Appl. Phys.* **98** 073301
- [5] Stoffels W W, Kemps P C M, Beckers J, Kroesen G M W and Haverlag M 2005 *Appl. Phys. Lett.* **87** 241501
- [6] Nimalasuriya T, Flikweert A J, Haverlag M, Kemps P, Kroesen G M W, Stoffels W W and Van der Mullen J A M 2006 *J. Phys. D: Appl. Phys.* **39** 2993
- [7] Flikweert A J, Van Kemenade M, Nimalasuriya T, Haverlag M, Kroesen G M W and Stoffels W W 2006 *J. Phys. D: Appl. Phys.* **39** 1599
- [8] Stoffels W W, Flikweert A J, Nimalasuriya T, Van der Mullen J J A M, Kroesen G M W and Haverlag M 2006 *Pure Appl. Chem.* **78** 1239
- [9] Stoffels W W, Van den Hout F J, Kroesen G M W, Haverlag M and Keyser R 2006 *Appl. Phys. Lett.* **88** 91502
- [10] Nimalasuriya T, Flikweert A J, Stoffels W W, Haverlag M, Van der Mullen J J A M and Papat N B M 2006 *J. Appl. Phys.* **99** 53302
- [11] Stoffels W W, Baede A H F M, Van der Mullen J J A M, Haverlag M and Zissis G 2006 *Meas. Sci. Technol.* **17** N67
- [12] Beks M L, Hartgers A and Van der Mullen J J A M 2006 *J. Phys. D: Appl. Phys.* **39** 4407
- [13] Nimalasuriya T, Thube G M, Flikweert A J, Haverlag M, Kroesen G M W, Stoffels W W and Van der Mullen J J A M 2007 *J. Phys. D: Appl. Phys.* **40** 2839
- [14] Flikweert A J, Nimalasuriya T, Kroesen G M W and Stoffels W W 2007 *Plasma Sources Sci. Technol.* **16** 606
- [15] Fischer E 1976 *J. Appl. Phys.* **47** 2954
- [16] Jinno M, Kurokawa H and Aono M 1999 *Japan. J. Appl. Phys.* **38** 4608
- [17] Jinno M, Takubo S, Hazata Y, Kitsinelis S and Motomura H 2005 *J. Phys. D: Appl. Phys.* **38** 3312
- [18] Vankan P, Antonis P and de Graaf A 2006 *Bull. Am. Phys. Soc. (GEC)* SRP2.00022
- [19] Sommerer T 2006 *Bull. Am. Phys. Soc. (GEC)* RR2.00005
- [20] Smith D J, Michael J D, Midha V, Cotzas G M and Sommerer T J 2007 *J. Phys. D: Appl. Phys.* **40** 3842
- [21] Yang X, Lin M, Zou W, Li Y and Zhang B 2003 *Phys. Chem. Chem. Phys.* **5** 4786