Molecular discharge lamps

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Abstract: A novel scheme for low pressure discharge lamps is explored. Rather than the commonly used mercury based fluorescent lamps, the use of molecular radiators emitting light in the UV or directly in the visible region is proposed. A lamp based on InBr is shown to emit light in the desired wavelength regime. LIF measurements, revealing the molecular and atomic kinetics, help to understand the underlying discharge mechanisms.

Keywords: Discharge lamps, LIF, molecules

1. Introduction

Currently lighting consumes about 20% of the worldwide electricity consumption. Approximately half of this light is produced by low pressure gas discharge lamps: the well known fluorescent tubes in all offices. The Hg in these lamps emits at 254 and 185 nm with an efficiency of about 70%. Unfortunately half of the photon energy is lost by the conversion of the UV photons to visible light by the luminescent phosphors on the wall. In addition Hg is a poisonous heavy metal. Therefore novel Hg-free discharge chemistries are considered that emit light in the near UV or directly visible region.

2. Discharge chemistry

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 [1]. Simple molecular gases like N_2 and H_20 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 [2]. It is known that metals are very efficient radiators, but in general their vapor pressure is too low at typical low pressure conditions. Exceptions are mercury and sodium. The latter is an efficient radiator, but mainly radiates on the well known orange lines. Sodium lamps are widely used in outside illumination, as the sodium emission lines match well with the optimum in the human eye response. However the color rendering is extremely poor, so it is not a viable lamp for indoor use, where white light is required. In high pressure lamps metal halide salts are used. Their efficiency is very good and white light can be obtained by mixing several salts. A significant partial pressure is obtained as the temperature in a high pressure lamp is above 1000K. In low pressure lamps the operating temperature is at most 400 to 500 K, so the vapor pressure of commonly used salts is virtually zero. The challenge is therefore to find metal containing molecules that have excellent radiating properties and having a low vapor pressure.

Indium-halides like InI and InBr are interesting candidates as they reach significant densities at relatively low pressures [3,4]. In this contribution we focus on the chemistry of InBr discharges. The spectrum of InBr (Fig 1) exhibits strong atomic 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. In this work we will look at the behavior of the plasma and using LIF we determine the plasma kinetics resulting in the spectrum shown in fig 1.

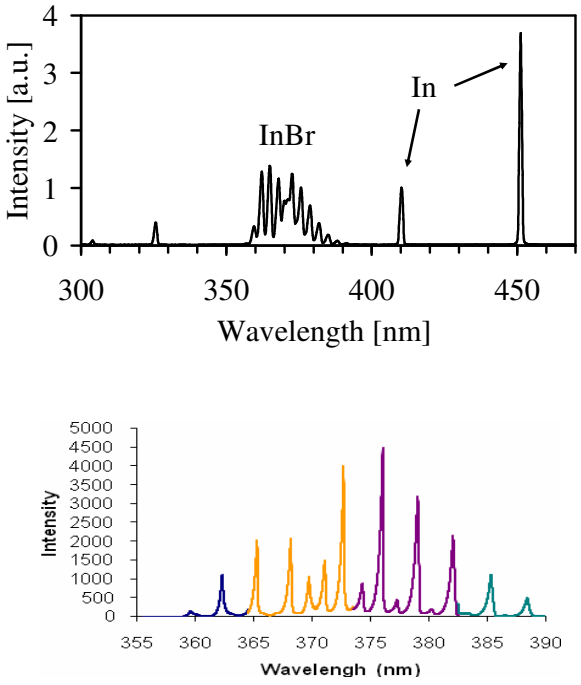

Fig. 1 Emission spectrum of InBr showing atomic lines at 410 nm and 450 nm as well as a molecular band. The lower picture shows a high resolution spectrum of the molecular band.

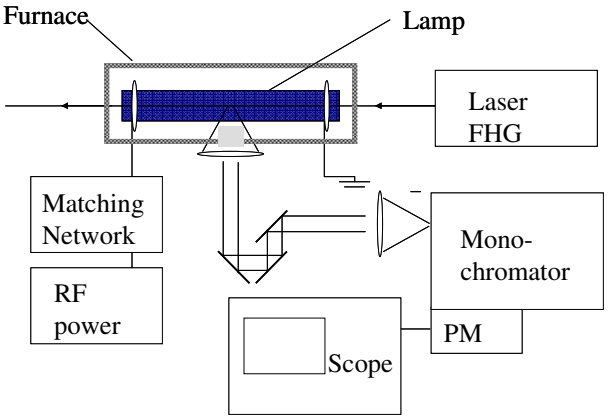

Fig. 2 A scheme of the experimental setup.

2. Experimental setup

A discharge is produced inside a sealed quartz cylinder (300 mm long, 38 mm diameter). The lamp is filled with argon as a buffer gas an 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 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. Figure 2 shows a simplified overview of the discharge setup including some diagnostics.

The discharge is studied spectroscopically using a spectrograph equipped with a photomultiplier and a CCD camera. Yang et al [5] 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 20 Hz 266nm Nd-Yag laser to excite the InBr. The integrated as well as the spectroscopically and time resolved responses are monitored.

2. Experimental results

At room temperature the InBr vapor pressure is very low and effectively an argon discharge is ignited. It is interesting to note that the discharge is prone to instabilities, which can result in spatially confined stable striation-like structures (figure 3)[6]. Several authors have reported similar structures, but a full explanation of their origin is not yet available. By increasing the temperature in the discharge, the vapor pressure of InBr increases. Above 450 K, the discharge changes color and effectively becomes a InBr discharge. Its spectrum is shown in fig 1. Under these conditions no argon emission lines are observed.

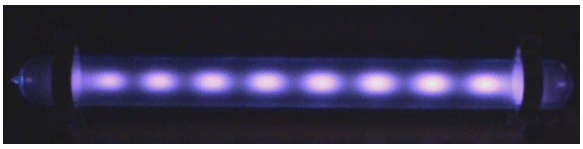

Fig. 3 A picture of the lamp at room temperature. Under these conditions stable geometric structures are easily formed.

The discharge is driven by a sinusoidal voltage on the electrodes. Using a Plasma Impedance Monitor we have verified the capacitive character of the discharge by monitoring the phase resolved current, as shown in figure 4. The phase angle between voltage and current is about 85°. Moreover the non-sinusoidal character of the current shows that there are various harmonics and non-linearities introduced by the discharge.

Figure 5 shows the energy potential diagram of InBr. 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 en 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. As can be derived from the energy potential diagram (fig. 5) two photons will cause photo dissociation of InBr and excited In is produced. This can be observed as resonant emission at 410 and 451 nm. An example of the 2-photon LIF signal is shown in figure 6.

In figure 7 the observed molecular laser induced fluorescence spectrum in the 360-390 nm range is shown. The 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 [5]. The general peak-structure is similar to the emission spectrum shown in figure 1. But both the relative magnitude of the various peaks as well as their shape show significant deviations. These differences contain the fingerprint of the coupling mechanisms between the various levels.

Figure 8 shows the time dependent behavior of the lif signal at 364.5 nm (0-0 B-X transition). As compared to the atomic lif data (figure 6) it is clear that both the rise time as the decay time of the molecular response is much slower. Furthermore, it is interesting to note that there is a large difference in the temporal behavior when the plasma is switched on. In presence of a plasma the lif

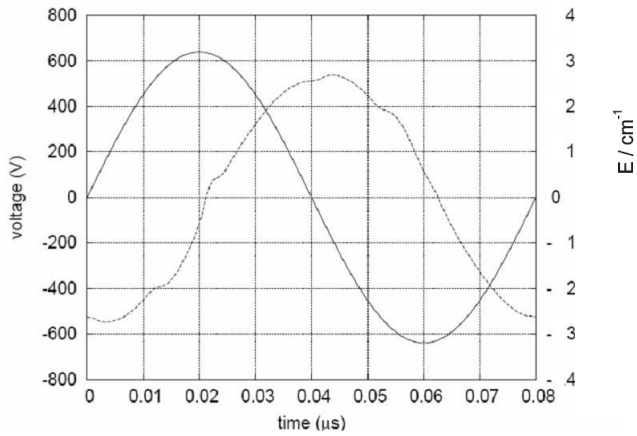

Fig. 4 Phase resolved voltage and current measurement at the electrodes of the discharge.

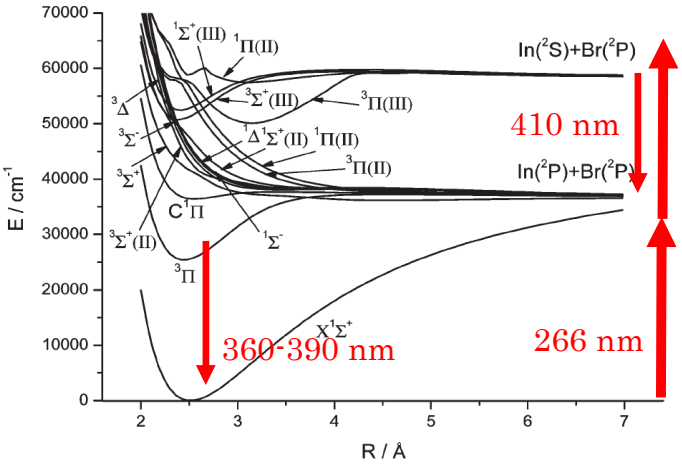

Fig. 5 Energy potential diagram of InBr (after [5]) together with the relevant laser (266 nm) and emission transitions and corresponding wavelengths (in red).

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.

Acknowledgements

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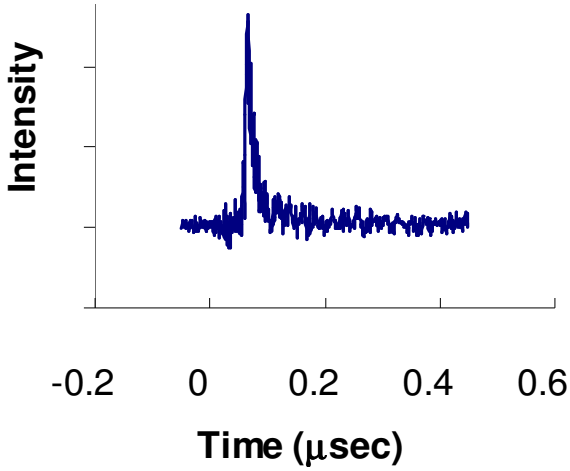

Figure 6, Laser induced fluorescence signal at 410 nm (atomic indium 2S-2P transition cf fig 5). The signal is generated in a heated InBr vapor at 280^{0}C (no plasma) in an argon buffer gas by two photon excitation using a 266 nm laser. The signal is recorded by a photomultiplier using a 1 m monochromator.

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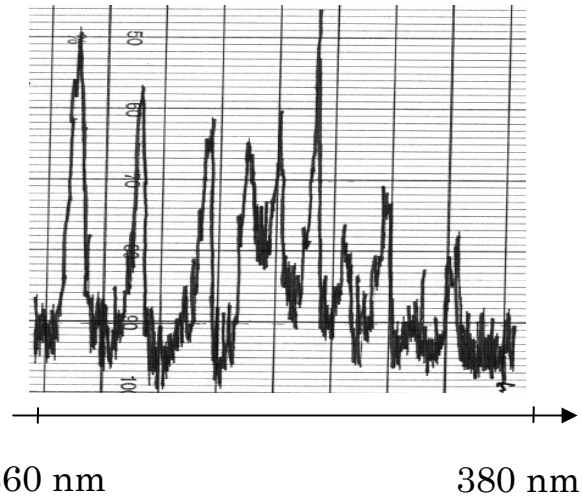

Figure 7, The 266 nm laser induced fluorescence signal of InBr observed between 360 and 380 nm (B-X and A-X transitions). The data are recorded by a photomultiplier, while scanning a 1 m monochromator. InBr is evaporated at 280^0C.

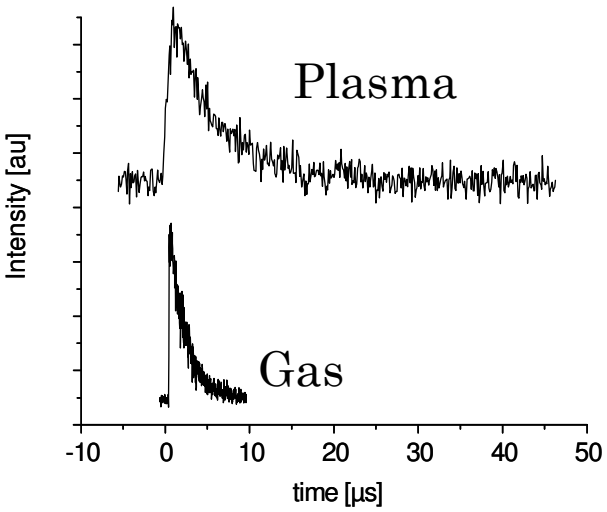

Figure 8, The time dependent 266 nm laser induced fluorescence signal of InBr observed at 364.52 nm (B-X $\Delta v = 0$ transition). With (top) and without plasma (bottom). The amplitude of the signals is scaled and shifted.