Implementation of a cold spot setup for controlled variation of vapour pressures and its application to InBr

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Discharges containing indium halides are discussed as efficient alternative for mercury based low pressure discharge lamps. To allow for a systematic investigation of the plasma properties a controlled variation of the indium halide density i. e. of the vapour pressure is mandatory. This can be achieved by utilizing a newly designed setup where a well-defined cold spot location is realized and the cold spot temperature can be adjusted between 50 and 350 °C. The performance of the setup has been verified by comparing the calculated evaporated InBr density (using the vapour pressure curve at the chosen cold spot temperature) with the one measured via white light absorption spectroscopy.

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I. INTRODUCTION

The utilization of indium halides as radiator in low pressure discharges for lighting applications is discussed as efficient alternative to common fluorescent lamps which contain hazardous mercury¹. Investigations^{2,3} revealed that the desired near-UV emission is strongly dependent on the evaporated indium halide amount which is determined by the temperature of the coldest spot of the discharge vessel wall T_{CS} . However, simply heating up the whole vessel to vary the indium halide density does not work properly as the position of the cold spot is not well-defined. Hence, increasing the heating temperature may also lead to a change of the cold spot location instead of changing the cold spot temperature. Therefore a well-defined cold spot position and the possibility to adjust T_{CS} is indispensable for systematic investigations. Thus a setup which fulfils these requirements has been developed.

In the setup presented in this paper, the whole vessel is heated up via a hot air blower but one defined spot of the vessel wall, the cold spot, is actively kept cooler. A variation of the cooling leads to a change of T_{CS} and therefore of the evaporated indium halide amount. This approach also has the advantage that the indium halide amount can be varied without changing the gas temperature of the plasma⁴. By varying the heating temperature of the whole vessel, both parameters are always changed simultaneously.

The performance of the well-defined cold spot setup is demonstrated at a sealed vessel filled with InBr salt. The InBr density values calculated from the adjusted value

II. COLD SPOT SETUP

A sketch of the setup is shown in figure 1. The discharge vessel (length 18 cm, diameter 2.5 cm) is actively heated via a hot air blower. The temperature of the hot air can be varied between ambient temperature and 600 °C. To achieve a homogeneous heating of the vessel, it is placed inside a heat container which is made out of autoclaved aerated concrete. To define the cold spot a copper plunger (diameter 8 mm) is connected via a graphite based high-temperature thermal conductance paste to the discharge vessel. This paste has to be renewed frequently to avoid degradation of the thermal contact. In order to enlarge the connection area, the surface of the copper plunger has a concave shape with the same radius as the discharge vessel. The temperature of the cold spot T_{CS} [°C] is measured by a type K thermocouple that is placed in a small groove between the plunger and the vessel. A heat pipe (diameter 6 mm, length 150 mm, pipe material copper, working fluid water) is used to assure a thermal connection between the

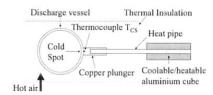


FIG. 1. Sketch of the cold spot setup. The cold spot temperature T_{CS} can be adjusted by heating or cooling the aluminium cube.

of T_{CS} and the vapour pressure curve are compared to those measured via white light absorption spectroscopy at different cold spot temperatures.

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copper plunger and an aluminium cube that is placed outside the thermal insulation. By actively adjusting the temperature of the aluminium cube the cold spot temperature can be set. For this reason, the cube can either be cooled by a thermoelectric cooler or heated by a heating wire. Heating the cube is necessary if higher cold spot temperatures than allowed by intrinsic cooling of the aluminium cube via convection are desired. The dimensions of the aluminium cube are rather large $(50 \times 50 \times 25 \text{ mm}^3)$ to slow down the thermal response time to a few minutes which stabilizes the cold spot temperature. With the described setup cold spot temperatures between 3 and 350 °C for measurements in gas phase can be adjusted. If a discharge is operated inside the vessel, the plasma heats the wall which limits the adjustable cold spot temperature range to 50 - 350 °C.

It is important that the temperature adjusted with the cold spot setup really represents the coldest temperature of the vessel wall. Therefore it is mandatory that the temperature of the hot air heating is considerably above the adjusted cold spot temperature. For example in the range of 150 °C < T_{CS} < 350 °C the heating temperature must exceed the cold spot temperature by more than 100 °C to assure a correct operation of the cold spot setup.

III. APPLICATION TO INDIUMBROMIDE

A. Determination of the ground state density of InBr via white light absorption spectroscopy

If light passes through a homogeneously absorbing medium of length l the transmitted intensity is given by⁵

$$I(\lambda, l) = I(\lambda, 0) \exp[-\tau(\lambda)],$$
 (1)

where $I(\lambda, x)$ is the intensity at wavelength λ and position x with $0 \le x \le l$. The optical depth $\tau(\lambda)$ of an absorption line arising from the transition $k \to i$ is defined as⁵:

$$\tau(\lambda) = \frac{\lambda_0^4}{8\pi c} \frac{g_i}{g_k} A_{ik} n_k \ l \ P_{line}(\lambda), \tag{2}$$

with the central wavelength of the absorption line λ_0 , the velocity of light in vacuum c, the statistical weights of the upper and lower state g_i and g_k , the transition probability for spontaneous emission A_{ik} , the density of the lower state n_k and the line profile P_{line} . By integrating the logarithm of the intensity over the wavelength of the absorption line, n_k can be determined:

$$n_k = \frac{8\pi c}{\lambda_0^4} \frac{g_k}{g_i} \frac{1}{A_{ik}l} \int_{line} \ln\left(\frac{I(\lambda, 0)}{I(\lambda, l)}\right) d\lambda.$$
 (3)

The absorption method can also be applied to electronic transitions of molecules. If the relative vibrational population $n_{i,rel}^{v'}$ of the upper electronic state i

is known, the population density of the whole state i can be derived using effective transition probabilities $^6A_{ik}^{eff}$. These can be evaluated by weighting the vibrationally resolved transition probabilities $A_{ik}^{v'v'}$ according to the relative vibrational population $n_{i,\ rel}^{v'}$ which is normalized to $\sum_{v'} n_{i,\ rel}^{v'} = 1$:

$$A_{ik}^{eff} = \sum_{v'} n_{i, rel}^{v'} A_{ik}^{v'v''}.$$
 (4)

In order to determine the density n_k of the lower electronic molecular state, the absorption signal has to be integrated over the whole wavelength range $\Delta\lambda$ of the electronic transition $k \to i$:

$$n_k = \frac{8\pi c}{\lambda_0^4} \frac{g_k}{g_i} \frac{1}{A_{ik}^{eff} l} \int_{\Delta \lambda} \ln \left(\frac{I(\lambda, 0)}{I(\lambda, l)} \right) d\lambda.$$
 (5)

For InBr, the absorption signal of the transition between the ground state X^1 $\Sigma^+(0^+)$ and the excited state B $^3\Pi_1(1)$ is measured. The corresponding spectrum is located in the near-UV spectral range between 350 and 400 nm. The required vibrationally resolved transition probabilities are taken from calculations which have been published previously⁷. As the measurements are carried out in gas phase, the population of the single vibrational states is calculated assuming a Boltzmann distribution according to the heating temperature.

B. Experimental setup

Figure 2 shows a sketch of the experimental setup used to check the performance of the cold spot. The vessel in which InBr is evaporated has a length of 18 cm and a diameter of 2.5 cm (wall thickness 2 mm). It is filled with 1.5 mg InBr salt and 1 mbar krypton as background gas. To provide access for white light absorption measurements at an axial line of sight (LOS) quartz windows in optical quality have been placed in the heat container. A stabilized high pressure xenon discharge lamp has been used as light source for absorption spectroscopy. The absorption spectra are recorded using a lens tube (aperture

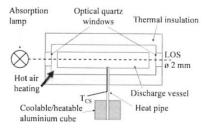


FIG. 2. Sketch of the experimental setup utilized for the performance test of the cold spot.

stop 2 mm) that collimates the transmitted light into an optical fibre leading to a high resolution spectrometer. The full width at half maximum of the Gaussian apparatus profile of the spectrometer at 350 nm is 23 pm.

C. Results

The vessel containing InBr and krypton was heated up to 500 °C. As the absorption signal of the InBr band is below the detection limit for $T_{CS} < 235$ °C the cold spot temperature has been varied between 250 and 305 °C. Figure 3 shows the comparison between the ground state density derived from absorption measurements and the InBr density calculated with T_{CS} , the ideal gas law (assuming the gas temperature equals the heating temperature) and the vapour pressure curve⁸. As expected from a well-defined cold spot setup, increasing T_{CS} results in an increase of the measured density. It can be seen that the InBr densities obtained with the two methods agree within the measurement errors. However, as the measured densities are slightly shifted to higher values it can be estimated that the adjusted T_{CS} -value which is measured at the outside wall of the discharge vessel is about 5 °C lower than the temperature of the actual cold spot which forms at the inner wall. This difference which is attributed to the occurrence of a temperature gradient across the vessel wall is indicated by error bars in figure 3.

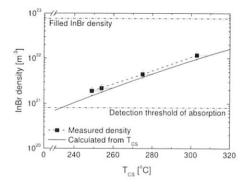


FIG. 3. InBr density measured with absorption spectroscopy compared to the density calculated after the vapour pressure curve.

IV. SUMMARY

For systematic investigations of indium halide containing low pressure discharges for lighting purposes a well-defined and adjustable cold spot at the discharge vessel wall is mandatory. This can be achieved by utilizing the presented setup. Its performance has been verified by white light absorption measurements of InBr. The measured densities agree well with those derived from the chosen cold spot temperature and the vapour pressure curve. It has been demonstrated that a controlled adjustment and variation of the evaporated InBr density can be performed with this cold spot setup.

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