Time-resolved spectroscopy of low-pressure discharges

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Abstract. Optical emission spectroscopy is used to investigate the excitation mechanisms in fluorescent tube plasmas. The temporal evolution of the intensities in the non-equilibrium parts of a pulse-excited rare gas – Hg mixture is recorded. Different transitions in a specific atomic system, as well as transitions from upper level of comparable excitation energy in different species, show distinctly different intensity build-up at the onset of the excitation, as well as varying decay characteristics in the afterglow after turning the excitation off. This implies different mechanisms for populating the excited level. The work of modelling the observations is in progress but hampered by the lack of adequate data for many of the important processes.

1. Introduction
While the general behaviour of the main discharge of a fluorescent tube (the “positive column”) is reasonably well understood [1], the same is not true for the processes in the vicinity of the electrodes, although the main features have been successfully modeled [2]. Electrodes are by design kept at a temperature between 1100 K and 1350 K in order to provide sufficient thermo-ionic emission to support the desired discharge current. A higher temperature causes excessive loss of emitter material due to evaporation, while a too low temperature, not sufficient to provide the necessary number of thermal electrons to maintain the current, causes a steep potential gradient to develop just outside the cathode. This potential step, the cathode fall, will accelerate mercury and rare-gas ions towards the cathode, and if their energy is sufficiently high these ions will cause sputtering of the emitter material, eventually leading to failure of the fluorescent tube.

During full-power operation, a suitable choice of rare-gas mixture and pressure and ohmic resistance of the electrode coils keeps the electrode temperature within the allowed limits. During ignition and during operation with reduced power (dimming), additional measures have to be taken to prevent destruction and shortened lifetime of the electrodes due to sputtering. Before applying the ignition voltage pulse to the tube, the electrode coils are therefore fed with for a short time with an auxiliary current. In the case of dimmed operation, this heating current has to be present all the time, and adjusted according to the main discharge current to maintain a suitable temperature. The power used for this auxiliary electrode heating is of the same order of magnitude as that used for light production when the tube is dimmed to low levels.

It seems clear [3,4] that a large fraction of the emitter material evaporated is subsequently ionized and pulled back to the electrode by the electric field during the cathode phase. This process is therefore of direct importance to the lifetime of a fluorescent tube. It has also been shown [5], that Penning processes are responsible for a large part of the ionization of mercury and most likely also
emitter material atoms. The present investigation was initiated by the desire to better understand the excitation/ionization mechanisms in the cathode region, especially concerning the emitter material, and to measure the dependence of these processes on mercury and the rare-gas mixture and pressure.

2. Experiment

The experimental setup is shown in Fig 1. A discharge tube with the fluorescent powder partially removed from its ends is driven by a broad-band (1MHz) A400DI power amplifier from FLC Electronics AB. The tube used in these measurements is filled with 99% argon and about 1% krypton, and a few mg of metallic mercury. The excitation waveform may be synthesized with components up to about 60 MHz, with an amplitude up to +/- 200 V.

The tube envelope is made of glass, preventing observation of ultraviolet (UV) lines. The exciting waveform in these measurements is symmetric, thus preventing axial cataphoresis during long runs. Data are collected in time windows synchronized to the exciting waveform. A microprocessor system is the heart of the data collection, controlling the waveform creation, aperture window timing, gating of the image intensifier in front of the CCD as well as that of the mechanical shutter of the spectrometer. Parameters to the microprocessor are transmitted from a MATLAB script running in a PC. After collecting data during a preset number of cycles, the script advances the time-window to the next point in the excitation cycle and repeats the procedure.

The light is analyzed by a 1 m Jarrel-Ash Czerny-Turner spectrometer and imaged onto a 1024x1024 pixel array CCD in an Andor DH534 ICCD camera head, equipped with a gateable image intensifier with a rise/fall time of about 50 ns. This sets the ultimate time resolution of the detection, but the overall bandwidth of the setup is limited by the power band-width of the amplifier to about...
1 MHz. Provisions are made for the simultaneous collection of spectra from six fibres. At each measurement, the full 1024 x 1024 pixel array image is saved. The channels of each spectrum are binned later during data evaluation, thereby allowing for the removal of spikes and “hot” channels, which otherwise often tend to severely distort the result.

3. Results

Figures 2 to 4 show recordings of the emission as a function of time. In all cases, the excitation wave form consisted of positive and negative moving pulses, separated by an idle time with no voltage applied to the electrodes. The length of the idle time periods, as well as the excitation pulse length is larger than the time scale of the plots shown. For all transitions shown here, the natural lifetime of the upper level is too small to be noticed on the plots. The electric field is in all cases turned off at t=0.

The energy of the upper $7s^3S_1$ level of the 4358 transition in Hg I is about 7.7 eV, very close to the upper level of the Ba II $\lambda$4554 transition shown in Fig 4. The Sr I emission shown in Fig. 3, however, only requires some 2.7 eV to be excited, but nevertheless shows definite similarity to the Ba II decay rather than to the Hg I decay. If, on the other hand, the excitation of the Ba II line is the result of Penning-type excitation from the ion ground state, rather than a single ionization-excitation event, then the energy available in this case, as well as for Sr I, is sufficient to populate extremely high-lying parts of the level system.

The observed decay in Figs. 3 and 4 may then be the result of cascading down a ladder of excited states, and the observed transition rate in this case cannot be possibly attributed to any individual process. Another possibility is that the observed slow decay of Figs. 3 and 4 reflects the depletion of rare-gas metastables, but in this case it remains to be explained why the metastables fail to excite the upper level of the Hg transition. It seems clear from the temporal behaviour of the fluorescent signal from the decay of the emission after turning off the electric field that the excitation of the emitter material atoms proceeds by multi-step processes, rather than by direct-electron excitation. A reasonable assumption is that Penning processes involving
rare-gas atoms in metastable states are responsible for a large fraction of the ionization/excitation of the emitter material [5].

The ionization of emitter material atoms during the cathode phase and subsequent “pulling back” of the ions to the electrode is probably a process of vital importance to the lifetime of the electrodes, and the investigation of this process as a function of the discharge parameters, especially the gas mixture, is therefore of great practical importance.

4. Conclusion
Information about the excitation channels may be deduced from the decay of the afterglow as well as from the rate of increase of emission intensities when the electrical excitation is turned on. Different transitions, sometimes even transitions in the same atom show different temporal behaviour. The explanation of this is subject of an on-going project, where the observations are matched to a simple one-dimensional model of the processes. This requires detailed knowledge of atomic and plasma parameters, such as excitation cross sections, electron energy distribution, excited state lifetimes, and charge transfer and Penning transition cross sections, many of which are currently poorly known. Work is in progress on such a model, initially containing some 10 neutral and ionized species, each with 10 to 100 excited states, using existing atomic data where available and reasonable guesses for the missing data.

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