5. Summary and outlook

In the frame of this study, it was achieved to investigate and describe the dynamics of single microplasma cavities in MSE array arrangements as well as their interaction under variation of operation parameters such as applied peak-to-peak voltage, excitation frequency, and gas pressure. This was realised by means of spectrally integrated two-dimensionally space and phase resolved optical emission spectroscopy using an ICCD camera and an attached long-distance microscope. The discharge mode of the investigated array devices was determined on the basis of voltage-current and photomultiplier tube measurements by comparison of observed behaviour to that of well-known dielectric barrier discharges in parallel plate geometry. The understanding of the processes responsible for single cavity dynamics and interaction is essential with respect to future tailoring of microplasma array devices to application demands and further scale-up.

The general comparability of the different MSE array devices was verified by voltage, current, and PMT measurements. All devices showed a synchronous self-pulsing of emission and current and comparable behaviour under variation of operation parameters. For each half period of the excitation period, a pulse train of current bursts was observed which is interrupted upon reversal of the applied voltage waveform slope.

In order to determine the discharge mode of the investigated devices, current peak behaviour under variation of operation parameters was examined. It was found, that with increasing excitation frequency the number of current peaks per pulse train decreases, their amplitude increases, and respective FWHM values decrease. It was thus discerned that at higher frequencies the number of long-lived excited species and surface charges originating from discharges in antecedent half periods is smaller so that higher overvoltages are required upon incipient burst onset. Consequently, less current peaks can evolve until the reversal of the slope of the applied voltage waveform. Typical derived current densities were in the order of 10 - 100 mA cm$^{-2}$. 
5. Summary and outlook

The self-pulsing was further investigated by means of complementary PMT measurements. It was observed that continuous emission builds up during a pulse train and superposes the single emission bursts. This behaviour is attributed to the successive build-up of metastable species as well as surface charge densities during a pulse train. These species can directly ionise nitrogen impurity species through Penning ionisation or cause secondary electron emission at the dielectric surfaces and effectively reduce ionisation threshold. For an increase of applied peak-to-peak voltage, the number of emission bursts per half period increased while the applied voltage upon incipient burst onset decreased. The FWHM value of subsequent bursts within one pulse train increased with increasing order of the burst, while the peak values decrease. This is further verification for the successive build-up of surface charge and metastable densities with successive emission bursts, which reduce the overvoltage upon ignition and can influence subsequent half periods.

The observed voltage, current, and emission behaviour in general agrees with that observed for parallel plate dielectric barrier discharges in pseudoglow mode, which is thus assumed for the investigated array devices as well. This has significant influence on the interpretation of experimental observations, as for every emission burst, a quasi-neutral glow discharge is formed. It accounts for substantially different discharge behaviour compared to Townsend-like discharges, which exhibit almost undisturbed electric fields.

Interaction of adjacent cavities in the array arrangements was investigated using two-dimensionally space and phase resolved optical emission spectroscopy. For this purpose, a recording and evaluation software was developed. It was observed, that each emission burst is composed of a successive ignition of the single cavities in the array arrangements in an isotropic wave-like manner. These ionisation waves were reproducible for the same operation parameters and did not depend on the locations of electric contacts on the array device. The starting point strongly depended on gas pressure, so that a preferred ignition due to pd-scaling resulting from manufacturing irregularities of single cavities was concluded. Typical propagation velocities were in the order of 1 - 10 km s\(^{-1}\).

In order to identify driving processes for ionisation wave propagation, experimental observations were complemented by a two-dimensional fluid simulation of a linear cavity arrangement. The simulation well reproduces the successive ignition of the adjacent cavities. The ionisation wave velocity resulting
from the simulation was 1 km s\(^{-1}\). The combination of experiment and simulation allowed to attribute ionisation wave propagation to electron and ion drift. Moreover, the simulation reproduced the formation of a glow discharge, which agrees well with experimental observations.

In the experiment, the ionisation wave propagation velocity increased with increasing excitation frequency. This increase is attributed to higher overvoltages upon ignition at higher excitation frequencies. Consequently, the electric field gradient upon ignition towards neighboured cavities under vacuum electric field is higher and may promote faster charge carrier drift velocities.

All of the investigated array devices were found to exhibit an asymmetry of applied voltage upon incipient burst onset as well as of emission intensity comparing both half periods of the excitation period. Registered emission intensity was typically smaller for negative Ni grid polarity while the polarity of earlier onset was dependent on the array design. As single cavity dynamics determine the collective array behaviour, it was examined by means of two-dimensionally space and phase resolved optical emission spectroscopy in order to investigate its impact on wave propagation and on asymmetric behaviour.

For all devices, the general behaviour of single cavity emission under variation of operation parameters agreed well with the observations made for integral devices. While the periodicity is the same, pulse widths are smaller for single cavities due to the wave-like superposition which is registered for integral array devices. However, conclusions drawn for integral devices were found to be valid for single cavities as well.

Emission intensity was found not to be homogeneously distributed within single cavities, but to exhibit peak structures close to the cavity edges. In this area, vacuum electric field strength is highest due to the stacking of the electrode structures. It was thus discerned that these peaked emission structures mainly originate from electron impact excitation. While for positive Ni grid polarity emission was registered up to 100 µm above the cavity and peaked closely to the cavity edges, it was concentrated inside the cavity and peaked more to the centre of the cavity for negative Ni grid polarity. This has significant influence on the interpretation of the observed asymmetric behaviour as the surface to volume ratio is smaller for positive Ni grid polarity. Thus, weighting of different processes deviates comparing both Ni grid polarities which can result in different plasma properties and the observed asymmetries.
Moreover, radiation transport as main driving process of ionisation waves can be excluded based on this observation. Peak emission at the cavity edges was only observed for cavities with a diameter bigger than 50 µm. For smaller cavity diameters, the peak structures superpose and build one effective central peak due to the proximity of opposite electrode edges.

Moreover, it was discerned that emission intensity in the cavity centre shows a significantly different temporal behaviour compared to that at the emission maxima. Both structures are superposed by an increasing continuous background and exhibit successively increasing FWHM values. While the peak intensity of the emission maxima decreases with increasing number of emission bursts per pulse train, that in the cavity centre remains constant. This indicates that substantially different excitation mechanisms prevail comparing emission maxima close to the cavity edges with the cavity centre.

Apart from the direct results presented before, this study illustrates the complexity of microplasma array systems due to single cavity and collective dynamics. The complicated diagnostic access due to geometric restrictions demands complementary simulations. However, the complex geometry and plasma chemistry of such MSE array devices cannot be fully implemented in current simulations due to lack of calculating capacity of present computers. Thus, simplifications of underlying models are required. In order to verify model assumptions, benchmark simulations, and verify processes responsible for the observed behaviour, further spectroscopic investigations are mandatory.

The results of this study suggest, that in particular the communication of subsequent half periods via long-lived species plays a major role in discharge dynamics. Here, metastable species and surface charges are most likely the main agents. If the drawback of interference of monoenergetic emission inside the ICCD camera and filter system can be resolved, this would yield huge application potential. It would allow a calibration of the system and thus phase and space resolved line-ratio measurements. In particular, in argon, the electron density [83, 84] and metastable densities [103] could be derived with temporal and spatial resolution.

Another way to temporally and spatially resolve argon metastable and argon ion densities on the dielectric surfaces and in the cavities could be provided by means of laser-induced fluorescence spectroscopy in combination with the ICCD camera filter system [104]. Ions could be excited from $3d^2D_9/2$ to $4p^2F_{7/2}$ at 661.5 nm and fluorescence to $4s^2D_{5/2}$ can then be observed at 459 nm [105].
5. Summary and outlook

Excitation of neutral argon metastables can be realised in the range of 690 - 790 nm. One realised scheme is based on excitation at 772.38 nm (1s₅ to 2p₇ in Paschen notation) and observation at 810.37 nm (2p₇ to 1s₄) [106]. Determination of absolute species densities is sophisticated due to the collisional operation regime. However, relative profiles and trends can yield valuable information on species behaviour.

Due to the restricted diagnostic access, conclusions on dominant processes in MSE array devices so far are based on variation of operation parameters. Further progress could be made with the availability of new device layouts. A variation of inter-cavity spacing and cavity aspect ratios could yield valuable information on intra- and inter-cavity processes. Further knowledge of such processes would then allow the design of new devices which may exploit the former in order to achieve high duty-cycles at low peak-to-peak voltages.

In certain applications such as high-frequency imaging with homogeneous lighting or lab-on-a-chip integration, where the interaction with single cavities cannot be readily monitored, the ionisation waves may be unfavourable. A re-design of MSE array devices with dielectric walls in between single cavities would not allow ion and electron drift between adjacent cavities. Thus, the propagation of ionisation waves may be prevented.