In this thesis, a cryogenic mechanically controllable break-junction (MCBJ) setup is developed and used for the characterization of single organic molecules at different temperatures. Molecules are the building blocks of life and among others responsible for charge transfer in biological processes, for instance photosynthesis in plants and metabolism in humans. Since these processes are not completely understood yet single molecules are highly interesting systems to study. Furthermore, due to their astonishing properties they might be used as electronic components in future technologies. This approach is called "molecular electronics", existing representatives are for instance light emitting diodes (OLED) or liquid crystal displays (LCD). However, single molecules devices are still not in use, and in order to build such systems the single molecules have to be understood, especially their electronic properties.

Since the size of single molecules is typically of the order of nm, their characterization requires an appropriate setup. In this thesis a MCBJ is developed that traps single molecules between two electrodes, and thus characterize its electronic features.

The first aim of this thesis is the construction and development of a cryogenic MCBJ setup, consisting mainly of three parts (i) the sophisticated sample holder, suitable for measurements inside the helium-flow cryostat, including a piezo-positioner to bend the sample (ii) an automated setup to control the measurement parameters and acquire the data (e.g. temperature, applied voltage, measured current, piezo-position), and (iii) the development of suitable break-junction samples consisting mainly of a lithographically prepared Au bridge (with nm size dimensions). The three parts allow measurements of molecules with a position control in the pm regime, currents ranging from below pA to mA, and temperatures ranging from 4 K to room temperature.

The second aim is the development and the establishing of adequate measurement procedures for break-junction measurements, regarding mounting of the sample, deposition of molecules and measurement techniques. For the latter one, conductance–position characteristics (CPCs), current–voltages characteristics (IVC) standard CPC histograms and contour histograms are developed, automated and tested in reference measurements without molecules.

The third aim is the measurement of simple "test-bed" molecules. Hexanedithiol and benzenedithiol are chosen as representatives for simple alkanes and conjugated molecules, respectively. CPCs of the rod-like hexanedithiol show a distinct peak in agreement with literature values, while BeDT exhibits more complex behavior. IVCs are performed on the molecules and molecular levels are obtained. The curves demonstrate the quality of the measurement techniques and the analysis methods.

Finally, the fourth aim is the measurement of the temperature dependent electronic properties of more complex molecules, terphenyldithiol (TPT) and porphyrine (TPyP). The latter one plays an important role in biological processes and represents a promising candidate for molecular electronics. TPT exhibits a distinct, and strongly temperature depended conductance peak, while the level of the molecular orbital is independent of temperature. It is shown that for TPT a transition from direct tunneling to "hopping" mechanism takes place around a temperature of 100 K. The TPyP displays unusual CPCs. Only tilted plateaus are observed, that are indicative for clustering of the molecule. Contour histograms demonstrate the presence of these clusters in the break-junction. IVCs of TPyP reveal a temperature dependent electronic or vibrational mode. It shifts towards higher voltages with increasing temperature and is therefore only detectable below $T \lesssim 180\,\mathrm{K}$. Above this temperature reordering of the molecules and the molecule-Au bonds lead to large noise in the IVC at large voltages.

The MCBJ represents an elegant way to detect interesting electronic properties of single molecules. Here, a major aspect is the great stability of the metal-molecule-metal system that allows extensive characterization even of complex molecules. In this thesis it is shown, that lower temperatures improve the performance of the MCBJ, and, even more crucial, temperature dependend measurements allow a deeper insight in the charge transfer of single molecules.