Abstract

The thesis at hand discusses synthesis and identification of previously unknown molecules and ions in the gas phase. To facilitate subsequent interstellar detection via radio astronomy, identification of the synthesized molecules and ions is accomplished, using laboratory rotational spectroscopy. The molecules and ions of interest are generated by means of radio frequency discharge. In addition to generation and *in situ* rotational spectroscopy, high level quantum chemical molecular calculations have to be carried out, in order to assign the measured rotational transitions. This work shows the viability of this approach.

At first it is demonstrated, that *in situ* rotational spectroscopy of discharge generated DSOD yields rotational transitions of the molecule, which can in turn be assigned on the basis of quantum chemical calculations carried out by *Prof. J. Gauß*. Altogether, more than 180 rotational transitions mainly of the ${}^{r}Q_0$-, ${}^{r}Q_2$- and ${}^{r}Q_3$-branches of DSOD in the frequency region between 75 and 655 GHz are detected and assigned.

Furthermore, two series of extensive quantum chemical calculations demonstrate the capability and accuracy of CCSD(T)/cc-p(C)VXZ (including anharmonic force field) calculations. Such calculations are carried out for the molecules SiS, H_2SiS, $(H_2SiS)_2$, HSN, HNS, and for the ions HS^+, H_2O^+, H_2S^+, $H_2S_2^+$. A comparison with available theoretical and especially experimental literature data shows the adequacy of these calculations as a basis for the assignment of measured rotational transitions.

At last, a systematic analysis of the radio frequency discharge of water and hydrogen sulfide demonstrates the potential and versatility of this method to generate unknown molecules and especially ions. This is accomplished by discharging water, hydrogen sulfide and a mixture of water and hydrogen sulfide (and their deuterated isotopomeres) respectively at different discharge energies and analyzing the generated plasma by means of a device known as Plasma Process Monitor. The resulting ion-mass spectra show a variety of partly unknown complex ions of interest like: $H_5O_2^+$, $H_7O_3^+$, $H_2S_x^+$, $H_3S_x^+$ up to $x = 5$.