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Outline and summary of this thesis

This thesis describes the design, development and progress of an experiment towards Doppler-free two-photon spectroscopy of the $(v, L) : (0, 3) \rightarrow (9, 3)$ ro-vibrational transition in HD^+ as proposed in Ref. [87]. Also some topics will be discussed which are closely related to the main experiment and were investigated by the author in the course of his tenure as a PhD student at Vrije Universiteit Amsterdam. The thesis is structured as follows:

Chapter 2: Recent advances in the *ab initio* calculations of energy level structure in hydrogen molecular ions are briefly mentioned. A brief summary of the measurement of the $v : 0 \rightarrow 8$ single-photon ro-vibrational transition in HD^+ is presented and its results are discussed [33, 86]. The experimental setup used for this measurement is briefly described as the Doppler-free two-photon spectroscopy experiment described in this thesis is largely built upon it. The systematic shifts and uncertainties of the $v : 0 \rightarrow 8$ measurement are presented, which explains that the 1.1 ppb fractional frequency uncertainty achieved in this experiment is limited almost exclusively by the first-order Doppler broadening. Also discussed briefly are the results of Ref. [87], which are essential for the design and realisation of the Doppler-free two-photon spectroscopy experiment.

Chapter 3: The experimental set up is described. Parts of the experimental set up were used to measure the $v : 0 \rightarrow 8$ vibrational overtone transition in HD^+ [33, 86] and have been described in more details elsewhere and hence discussed briefly. The spectroscopy laser system for probing the two-photon transition is extensively described together with its performance.

Chapter 4: The choice of hyperfine component of the $(v, L) : (0, 3) \rightarrow (9, 3)$ ro-vibrational transition to be measured is discussed on the basis of minimum net Zeeman shift and suppressed Doppler-broadened background, which are essential for a strong signal. An extended rate-equation model has been developed following the work of Biesheuvel *et al.* [33, 86] for modelling the expected two-photon spectrum. This model is useful for *a priori* determination of required experimental parameters such as appropriate laser detunings, optimum excitation duration and laser intensities, the experimental optimisation of which would be extremely tedious. The simulated two-photon lineshape can also be used to estimate shift in the estimation of the resonance frequency of the Doppler-free feature due to the Doppler-broadened background. The AC Stark shift induced by the dissociation laser at 532 nm and the cooling laser at 313 nm lasers as well as the Zeeman shift are discussed.

Chapter 5: "Ghost features" in Doppler-broadened ro-vibrational spectrum of trapped, laser-cooled HD^+ ions are discussed. These are features appearing in the Doppler-broadened single-photon spectrum in a finite sample of molecules (or atoms) with closely spaced components, the line profiles of which are par-

tially overlapping due to first-order Doppler broadening. The example case of the $(v, L) : (0, 3) \rightarrow (4, 2)$ single-photon ro-vibrational transition of HD^+ has been studied. The ghost features are found to appear in the spectrum where there are no underlying hyperfine components of the ro-vibrational transition. The conditions in which these ghost features appear have been studied and their dependence on experimental parameters such as translational temperature of the sample molecules, finite number of the sample molecules, laser intensities and probe durations are extensively discussed and their implications indicated.
Chapter 6: A re-derivation of the proton-electron mass ratio $\mu_{\rm pe}$ from the existing spectroscopic measurements of ro-vibrational transitions in HD^+ in the light of CODATA-14 prescribed set of fundamental constants is presented. This has important consequences for the determination of the same constant from the Doppler-free two-photon experiment.
Chapter 7: The first observation of the $(v = 9)$ vibrational level in the ground electronic state of HD^+ is reported. Recent progress towards the observation and measurement of the Doppler-free two-photon ro-vibrational transition in HD^+ is presented. Some evidence has been presented towards the observation of the $(v, L) : (0, 3) \rightarrow (9, 3)$ Doppler-free two-photon transition.
Chapter 8: The thesis is concluded with the discussion of some prospects of the Doppler-free two-photon experiment in HD^+.