DETERMINATION OF THE SPIN-ROTATION FINE STRUCTURE OF $He_2{}^+$

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Measuring spin-rotation intervals in molecular cations is challenging, particularly so when the ions do not have electric-dipole-allowed rovibrational transitions. We present a method to determine the spin-rotational fine structure of molecular ions from the fine structure of high Rydberg states[a]. The method is illustrated by the determination of the so far unknown spin-rotation fine structure of the fundamentally important $He_2{}^+$ ion in the $X^+\ {}^2\Sigma_u^+$ ground electronic state. The interaction that is responsible for the level structure in the high Rydberg states of He_2 that were probed in our experiment is the n-independent spin-rotation interaction of the ion core. As a consequence, the fine-structure splittings in $He_2{}^+$ can be related to the fine-structure of the Rydberg states by applying an angular-momentum basis transformation from Hund's case (e[b]) to Hund's case (d).

The experiment relies on the use of single-mode cw radiation to record spectra of high Rydberg states of He_2 from the a ${}^3\Sigma_u^+$ metastable state. Metastable helium molecules are produced by striking a discharge in a pulsed expansion of neat helium gas[b]. Cooling the valve body to a temperature of 10 K and using continuous-wave excitation results in an observed Doppler-limited linewidth of 25 MHz. The fine structure of Rydberg states of He_2 is determined from strict selection rules by comparing the observed splitting of the Rydberg spectrum with the spin-rotational intervals of the initial metastable state. The fine-structure splittings of the $v^+ = 0$, $N^+ = 1, 3$, and 5 levels of $He_2{}^+$ are 7.96(14)MHz, 17.91(32) MHz and 28.0(6) MHz, respectively.

[a]P. Jansen, L. Semeria, and F. Merkt, *Phys. Rev. Lett.* **120**, 043001 (2018).

[b]M. Motsch, P. Jansen, J.A. Agner, H. Schmutz, and F. Merkt, *Phys. Rev. A* **89**, 043420 (2014).