
Supplementary information

Proton–electron mass ratio by high-resolution optical spectroscopy of ion ensembles in the resolved-carrier regime

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**Supplementary Information for “Proton-electron mass ratio by
high-resolution optical spectroscopy of ion ensembles in the
resolved-carrier regime”**

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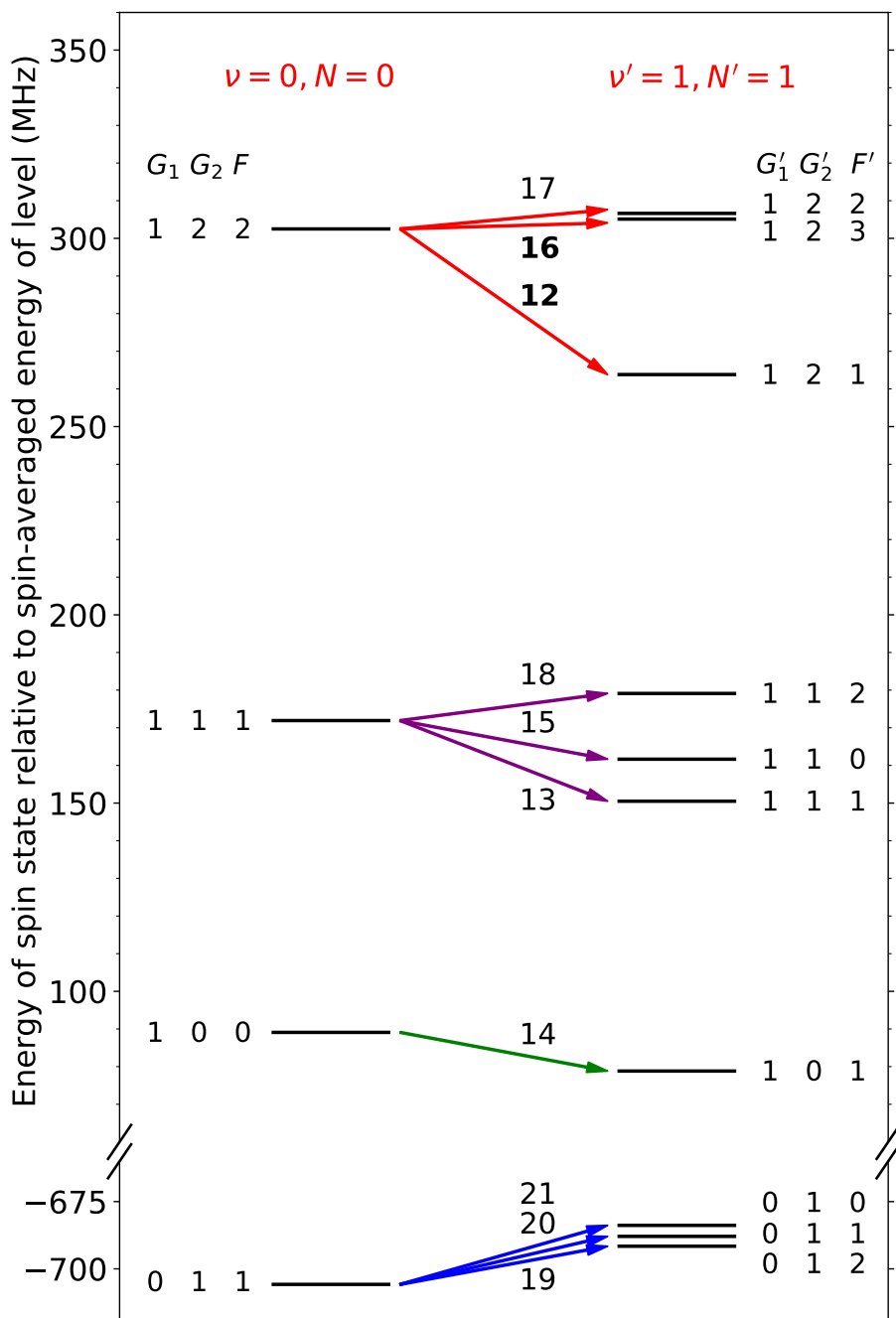
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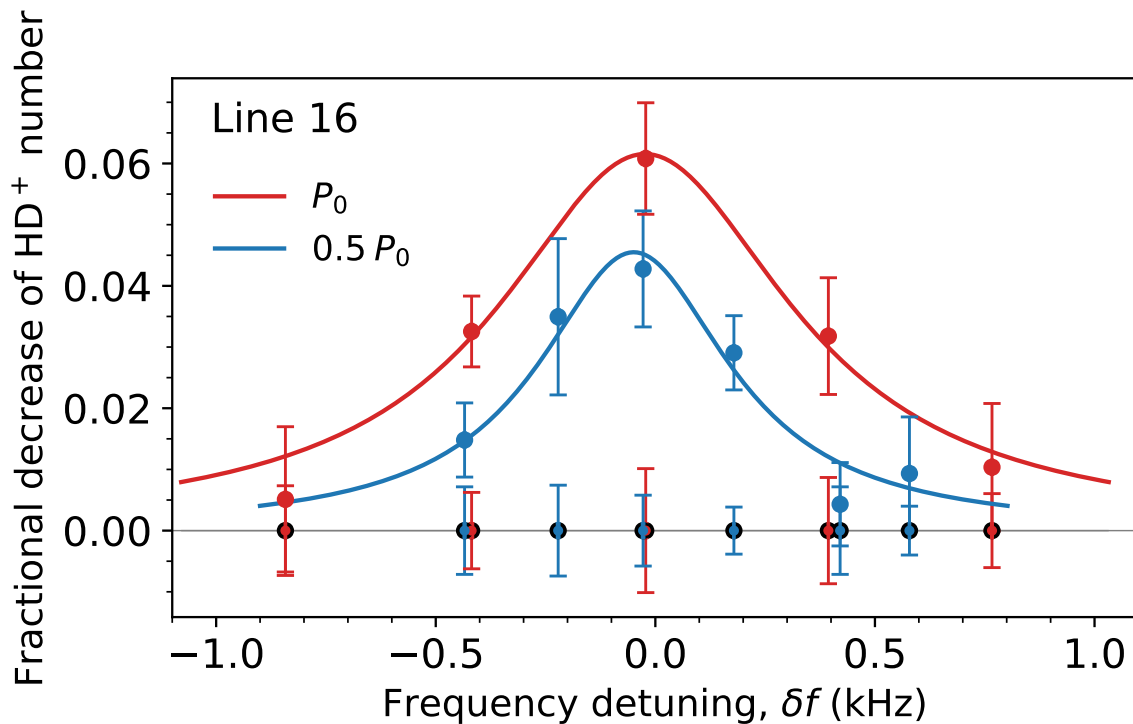
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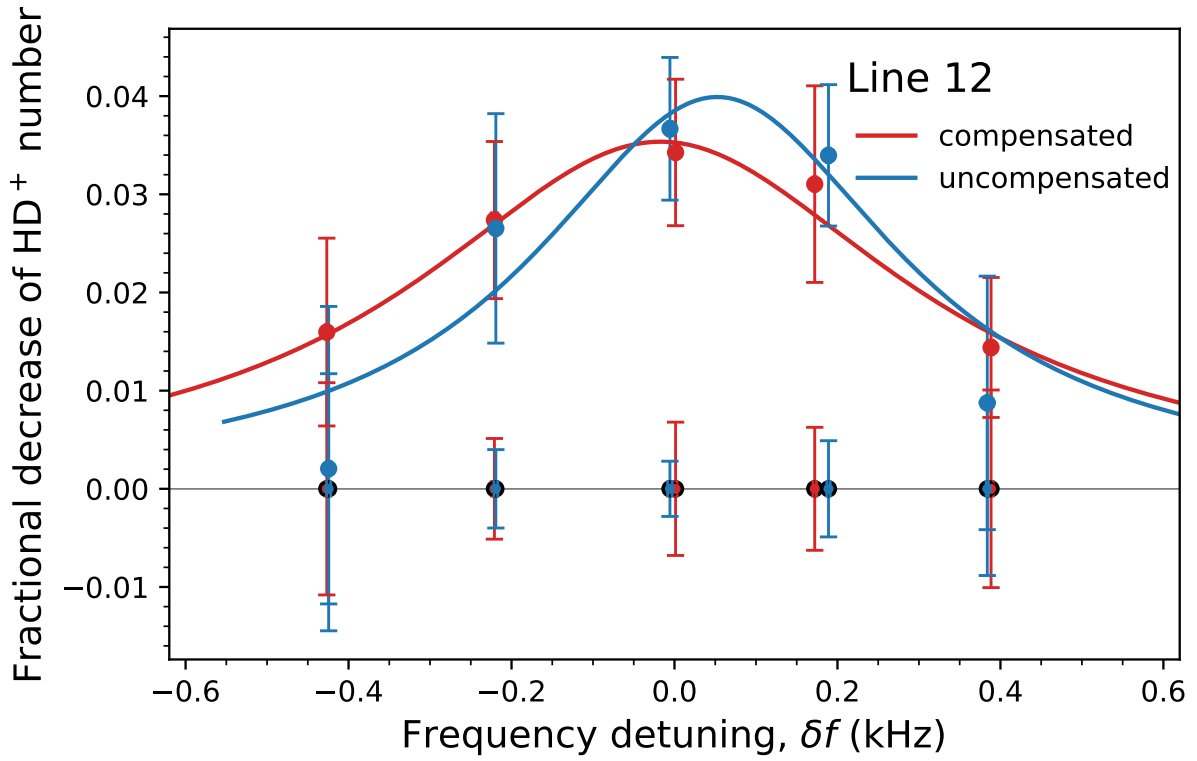
Supplementary Figure 1. **Hyperfine structure energy levels of HD⁺ in the two relevant levels.** Shown are the ground vibrational level ($\nu = 0, N = 0$) (left) and the first excited vibrational level ($\nu' = 1, N' = 1$) (right) of the $^2\Sigma_g^+$ electronic state. The numbers next to the arrows indicate the line numbers. In this work, lines 12 and 16 were detected and measured.

		\mathcal{E}'_1	\mathcal{E}'_2	\mathcal{E}'_3	\mathcal{E}'_4	\mathcal{E}'_5	\mathcal{E}'_6	\mathcal{E}'_7	\mathcal{E}'_8	\mathcal{E}'_9	\mathcal{E}_4	\mathcal{E}_5
		30.28083	-0.03046	-0.004664	903.36802	138.91049	8.13669	1.24894	-0.002945	0.005659	925.39588	142.28781
Line i	$f_{\text{spin},i}^{(\text{theor})}$	$\gamma'_{i,1}$	$\gamma'_{i,2}$	$\gamma'_{i,3}$	$\gamma'_{i,4}$	$\gamma'_{i,5}$	$\gamma'_{i,6}$	$\gamma'_{i,7}$	$\gamma'_{i,8}$	$\gamma'_{i,9}$	$\gamma_{i,4}$	$\gamma_{i,5}$
12	-38.68609	-0.575	-0.565	-1.715	0.250	0.430	-0.011	-3.369	-3.306	-2.909	0.250	0.500
16	2.60772	0.500	0.500	1.000	0.250	0.500	-0.500	-1.000	-1.000	-0.500	0.250	0.500

Supplementary Table 1. **Spin hamiltonian coefficients, spin structure frequencies, and spin frequency derivatives.** \mathcal{E}'_k are the coefficients of the spin Hamiltonian [45] for the $(v' = 1, N' = 1)$ level, in MHz. \mathcal{E}_k are the coefficients for the ro-vibrational ground state $(v = 0, N = 0)$, already reported in [13]. $f_{\text{spin}}^{(\text{theor})}$ is the theoretical spin structure frequency in MHz. γ are the dimensionless sensitivities of the spin structure frequencies to the various spin Hamiltonian coefficients. $\gamma'_{i,k} = \partial f_{\text{spin},i}^{(\text{theor})} / \partial \mathcal{E}'_k$ refers to the upper state, $\gamma_{i,k} = -\partial f_{\text{spin},i}^{(\text{theor})} / \partial \mathcal{E}_k$ to the lower state.



Supplementary Figure 2. **Power broadening of one Zeeman component of the vibrational transition.** The $m_F = 0 \rightarrow m'_F = 0$ component of line 16 was interrogated at two power settings of the $5.1 \mu\text{m}$ spectroscopy laser. Here, $B \simeq 0.6 \text{ G}$. The zero of the frequency detuning is arbitrary. P_0 is the nominal power used in the measurements shown in the main text. Each error bar represents the standard deviation of the mean.



Supplementary Figure 3. **The effect of a displacement of the beryllium ion cluster from the trap axis.** One Zeeman component of the vibrational transition is measured with the beryllium ion cluster aligned with the trap axis (red) and with the cluster significantly shifted radially (blue). The $m_F = 0 \rightarrow m'_F = 0$ component of line 12 is shown. The zero of the frequency detuning is arbitrary. Each error bar represents the standard deviation of the mean.