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# Characterization of a Static Magnetic Field with Two-Photon Rotational Spectroscopy of Cold Trapped HD<sup>+</sup>

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**Abstract**—Two-photon rotational spectroscopy of cold trapped HD<sup>+</sup> ions may be exploited for characterization of the magnitude and orientation of a static magnetic field in the ion trap. The experimental setup and the approach for calibration of the magnetic field vector are described. A sensitivity at the 10<sup>-11</sup> T level may be reached with Zeeman spectroscopy of a hyperfine component of the (v,L)=(0,0)→(0,2) transition. The orientation of a magnetic field with a magnitude at the 1 μT level may be characterized with an uncertainty better than 30 mrad.

**Keywords**—two-photon spectroscopy, trapped ions, hydrogen molecular ions, Zeeman shift, ab-initio calculation, vector magnetometry

## I. INTRODUCTION

Precise sensing of the magnetic fields has applications in geophysics [1], medical imaging [2,3], and precision physics measurements [4]. The atomic magnetometers exploiting alkali atoms vapours are extremely sensitive [5], but their response, based on Larmor precession, is intrinsically scalar. This work evaluates the potential for SI-traceable full characterization of a static magnetic field from precision measurements of transitions of trapped molecular ions compared with the predictions of the state-of-the-art ab-initio energy level calculations. Precisely, the energy levels of the hydrogen molecular ions (HMI), the simplest molecular quantum systems, were predicted with quantum electrodynamics (QED) ab-initio calculations reaching a fractional accuracy at the 10<sup>-12</sup> level [6]. The shifts of the energy levels due to various external fields were also predicted by ab-initio calculations [7,8]. Particularly, the effects of a static magnetic field on the HD<sup>+</sup> energy levels were evaluated in the nonrelativistic approximation at the order α<sup>2</sup>. The ab-initio calculations allowed identifying and exploiting for precision measurements transitions which are insensitive to the magnetic field and to its orientation with respect to the polarization of the radiation field. The rovibrational and rotational spectroscopy of trapped and sympathetically cooled HD<sup>+</sup> ions was performed by resonant-enhanced multiphoton dissociation (REMPD). The Doppler-limited infrared spectroscopy allowed a fractional uncertainty at the 10<sup>-9</sup> level [9]. A significant increase in the accuracy and resolution can be provided by the Doppler-free spectroscopy. The rotational spectroscopy of HD<sup>+</sup> in the Lamb-Dicke regime allowed kHz-level resolution and a fractional accuracy at the 10<sup>-10</sup> level [10]. A double-resonance two-photon spectroscopy scheme of HD<sup>+</sup> may provide fractional accuracy and resolution at the 10<sup>-12</sup> level or better [11].

In precision measurements, a static magnetic field, generated with a set of coils, allows to define the quantization axis of the molecular ions and to split their energy levels. This contribution proposes to exploit the scalar response obtained from the HD<sup>+</sup> ions by two-photon Zeeman spectroscopy to characterize the magnitude and orientation of the magnetic field.

## II. THEORY AND SPECTROSCOPY OF HD<sup>+</sup>

The rovibrational energy levels of HD<sup>+</sup> in its ground electronic state are calculated as the nonrelativistic Schrödinger energy plus a series expansion of QED correction terms [6]. At high resolution, the energy levels reveal a hyperfine structure described with the coupling of the proton  $\vec{I}_p$ , deuteron  $\vec{I}_d$ , and electron  $\vec{S}_e$  spins with the rotational angular momentum  $\vec{L}$  to yield  $\vec{J}$ , the total angular momentum:  $\vec{F} = \vec{S}_e + \vec{I}_p$ ;  $\vec{S} = \vec{F} + \vec{I}_d$ ;  $\vec{J} = \vec{L} + \vec{S}$ . The hyperfine eigenstates  $|vLFSJ\rangle$ , labelled with the quantum numbers for the vibration  $v$  and for the angular momentum coupling scheme, are calculated using a Breit-type spin Hamiltonian and accurate variational Coulomb wavefunctions [12]. A small magnetic field is used to define the quantization axis and to split the hyperfine states in magnetic subcomponents, labelled with the quantum number  $J_z$  of the projection of  $\vec{J}$  on the quantization axis. The Zeeman shift of an energy level in an external static magnetic field may be described with a quadratic dependence in function of the magnetic field magnitude [7]. The total energy of a magnetic level can be expressed as :

$$E(v,L,F,S,J,J_z) = E_{rv}(v,L) + E_{hf}(v,L,F,S,J) + \Delta E_z(v,L,F,S,J,J_z;B) \quad (1)$$

in function of the rovibrational energy  $E_{rv}(v,L)$ , the hyperfine energy  $E_{hf}(v,L,F,S,J)$ , and the Zeeman shift  $\Delta E_z(v,L,F,S,J,J_z;B)$ , respectively.

Accurate measurements of the HD<sup>+</sup> transitions can be performed with an experimental setup based on cold trapped ions [9,10]. Precisely,  $\sim 10^2$  HD<sup>+</sup> ions and  $\sim 10^3$  Be<sup>+</sup> ions are trapped together in a radiofrequency trap. The Be<sup>+</sup> ions are laser cooled with a 313 nm laser. The electrostatic interactions between two species allow sympathetical cooling of the HD<sup>+</sup> ions. When the secular motion of the HD<sup>+</sup> ions is excited in the trap, the increase in fluorescence

of the laser-cooled  $\text{Be}^+$  ions is used to monitor the number of the trapped  $\text{HD}^+$  ions. A static magnetic field, controlled with three coil pairs, driven with three independent current sources, is applied to the ion trap. The measurements proposed in this contribution are based on the double resonance spectroscopy scheme discussed in [11], that addresses the two-photon rotational transition in the vibrational ground state  $(v,L)=(0,0)\rightarrow(0,2)$  and the two-photon rovibrational transition  $(v,L)=(0,0)\rightarrow(2,0)$ . The detection is performed by the photodissociation of the  $(v,L)=(2,0)$  level with a 175 nm laser. The Doppler-free spectroscopy is performed with two counterpropagating waves tuned around the rovibrational transition at 55.909 THz, and the rotational transition at 1.968 THz, respectively (Fig. 1). The two-photon transition rates between the magnetic levels are derived using the two-photon operator formalism and the second-order time-dependent perturbation theory. The interaction with the blackbody radiation at room temperature recycles continuously the population among the rotational levels of the ground vibrational state. Moreover, the population in an excited energy level decays radiatively. The change of the population in the rovibrational levels is calculated with a set of rate equations in order to derive the lineshapes of the two-photon resonances [11]. The full-width half measured linewidth is 12.1 Hz for the rovibrational line and 0.447 Hz for the rotational line, by assuming a two-photon rotational transition rate of  $2000 \text{ s}^{-1}$ , a two-photon rovibrational transition rate of  $10 \text{ s}^{-1}$ , a dissociation rate of  $200 \text{ s}^{-1}$  and a REMP time of 10 s.

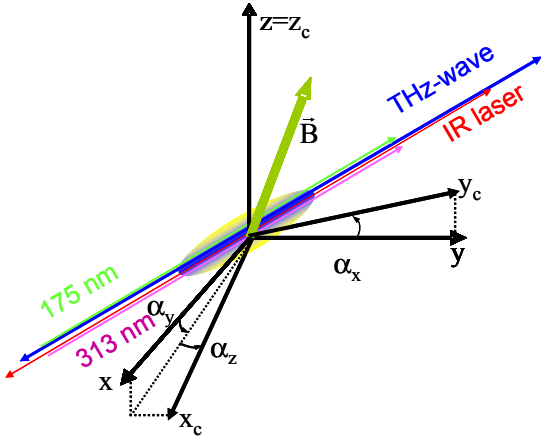


Fig. 1. The experimental setup and the reference coordinate frames.

The fractional frequency uncertainty is estimated with the Allan variance for the quantum projection noise limit :

$$\sigma_y(\tau) = \frac{1}{\pi Q \sqrt{N_{ion}}} \times \sqrt{\frac{T_c}{\tau}} \quad (2)$$

expressed in function of the quality factor of the two-photon transition  $Q = f_{2ph}/\Delta f_{HWHM}$  in terms of the half-linewidth determined previously. The cycle time  $T_c$  is associated to a single measurement with  $N_{ion}$  ions at the two-photon resonance. Successive measurements are averaged during an interrogation time  $\tau$ . Assuming the case of single ion spectroscopy experiment with  $T_c = \tau$ , the frequency uncertainty for  $(v,L)=(0,0)\rightarrow(2,0)$  line is estimated at 1.9 Hz and for  $(v,L)=(0,0)\rightarrow(0,2)$  line at 71 mHz.

The Zeeman shift of a transition  $|v,L,F,S,J,J_z\rangle \rightarrow |n',J'_z\rangle$  may be calculated approximatively with a quadratic dependence :

$$f_{n,n'}(B) - f_{n,n'}(B=0) = \eta_B(t_n, q_n, r_n, t_{n'}, q_{n'}, r_{n'}, J_z, J'_z) B + \eta_{B^2}(t_n, q_n, r_n, t_{n'}, q_{n'}, r_{n'}, J_z, J'_z) B^2 \quad (3)$$

in function of the magnitude of the magnetic field  $B$  and a set of six theoretical parameters  $\{U_{th}\} = \{t_n, q_n, r_n, t_{n'}, q_{n'}, r_{n'}\}$  that were calculated ab-initio for the relevant hyperfine energy levels of  $\text{HD}^+$  in [7]. The favoured (intense) two-photon electric dipole (E1-E1) transitions obey the selection rules  $\Delta F = \Delta S = 0$ ,  $\Delta J = 0, \pm 2$ . The linear and quadratic Zeeman shift coefficients are calculated and displayed in Table I for a selection of magnetic subcomponents of the rotational and rovibrational two-photon transitions. The subcomponents of the rotational transition, in general more sensitive than the subcomponents of the rovibrational transition, are suitable for magnetic field characterization. When the THz source used for two-photon spectroscopy is referenced to a frequency standard, this approach allows absolute calibration of the magnetic field in the ion trap, using Zeeman spectroscopy measurements, ab-initio calculations of the  $\text{HD}^+$  energy levels and of their Zeeman shifts.

TABLE I. ZEEMAN SHIFT COEFFICIENTS FOR SELECTED COMPONENTS OF THE TWO-PHOTON ROTATIONAL AND ROVIBRATIONAL TRANSITIONS.

$v,L,F,S,J,J_z$	$v',L',F',S',J',J'_z$	$\Delta\eta_B(\text{kHz/T})$	$\Delta\eta_{B^2}(\text{MHz/T}^2)$
0, 0, 1, 0, 0; 0	2, 0, 1, 0, 0; 0	0	40900
0, 0, 0, 1, 1; $\pm 1$	2, 0, 0, 1, 1; $\pm 1$	$\mp 155$	4700
0, 0, 1, 2, 2; $\mp 2$	0, 2, 1, 2, 4; 0	$\pm 6992320$	1855850
0, 0, 1, 1, 1; $\mp 1$	0, 2, 1, 1, 3; $\pm 1$	$\pm 6123100$	397050

### III. CHARACTERIZATION OF A STATIC MAGNETIC FIELD

This part will describe a procedure for measuring the magnitude and orientation of a static magnetic field in the Cartesian Laboratory Coordinate Frame  $LCF(\vec{e}_x, \vec{e}_y, \vec{e}_z)$ , by using two-photon Zeeman spectroscopy of  $\text{HD}^+$  (Fig. 1). The three coil pairs define a Coil Coordinate Frame  $CCF(\vec{e}_{c,x}, \vec{e}_{c,y}, \vec{e}_{c,z})$  which is not necessary orthogonal. The relative orientations of the  $CCF$  axis relative to  $LCF$  are defined with the Euler angles :  $(\alpha_z, \pi/2 + \alpha_y)$  for  $\vec{e}_{c,x}$ ,  $(\pi/2, \pi/2 - \alpha_x)$  for  $\vec{e}_{c,y}$ , and  $(0,0)$  for  $\vec{e}_{c,z}$ , respectively. A slight nonorthogonality of  $CCF$  is accounted with the small angles  $\alpha_x, \alpha_y, \alpha_z \ll 1$ .

The measurement of the Zeeman shift  $\mathcal{E}$  of a magnetic subcomponent of the two photon rotational transition  $|vLFSJJ_z\rangle \rightarrow |n,J'_z\rangle$  allows to calculate the magnitude of the magnetic field in function of the relevant values for the theoretical parameters  $\{U_{th}\}$ , by inverting eq. (3). A choice between the two values of the magnetic field derived from the second-order equation may be made by comparison with the value obtained with an accurate magnetometer. The uncertainty of the magnetic field may be calculated with the error propagation formula, in function of the contribution from the frequency shift measurement uncertainty

$\sigma_{exp,rot} = 0.1 \text{ Hz}$ , estimated with the quantum projection noise limit, and the contributions from the theoretical parameters, which are assumed correlated and having the same uncertainty  $\sigma_{th,q,r} = 50 \text{ MHz/T}^2$ ,  $\sigma_{th,t} = 5 \text{ kHz/T}$ . Here, the Zeeman spectroscopy of the subcomponent  $|v,L,F,S,J,J_z\rangle = |0,0,1,2,2,-2\rangle \rightarrow |0,2,1,2,4,0\rangle$ , having the highest Zeeman shift of  $717.791 \text{ kHz}$  at  $B = 10^{-4} \text{ T}$ , is exploited for measuring the magnetic field. The dependence of the uncertainty with the magnitude of the magnetic field in the ion trap is plotted in Figure 2. The sensitivity of the two-photon Zeeman spectroscopy method is given by the experimental uncertainty of  $1.4 \times 10^{-11} \text{ T}$ . Moreover, a magnetic field smaller than  $10^{-4} \text{ T}$  may be calibrated with a precision better than  $7 \times 10^{-11} \text{ T}$ .

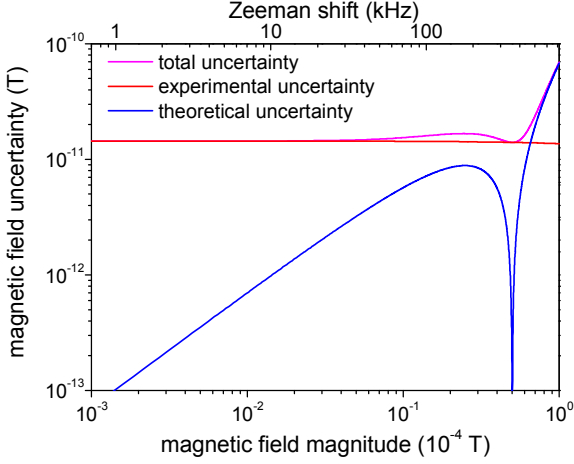


Fig. 2. Total uncertainty (magenta line), the contribution from the frequency measurement (red line), and the contribution from the theoretical calculation (blue line) of the magnitude of the magnetic field measured by Zeeman spectroscopy of the  $|v,L,F,S,J,J_z\rangle = |0,0,1,2,2,-2\rangle \rightarrow |0,2,1,2,4,0\rangle$  transition of  $\text{HD}^+$ . The frequency uncertainty is estimated with the quantum projection noise-limited instability for a single-ion experiment with integration time 1 s and cycle time 1 s.

The directional measurements of the magnetic field are determined by a set of experimental parameters: three currents ( $I_1, I_2, I_3$ ) that drive independently the coil pairs in Helmholtz configuration, the current-to-field parameters of the coil pairs ( $k_1, k_2, k_3$ ), three angles that define the relative orientation of the coil pairs ( $\alpha_x, \alpha_y, \alpha_z$ ), and the offset magnetic field components ( $B_{01}, B_{02}, B_{03}$ ), due for example to the Earth's magnetic field. As discussed previously, the magnitude of the magnetic field can be expressed in function of the experimental frequency of a two-photon transition  $f_{exp,i}$  shifted by the Zeeman effect from the theoretical value  $f_{th,i}$  calculated at zero-field in proportion with the linear Zeeman shift coefficient  $\eta_i$ :

$$\|\bar{B}(\{V_k\}_i; I_{1,i}, I_{2,i}, I_{3,i})\| = \frac{f_{exp,i} - f_{th,i}}{\eta_i} \quad (4)$$

This equation assumes linear approximation. The set of nine parameters  $\{V_k\}_i = \{k_1, k_2, k_3, \alpha_x, \alpha_y, \alpha_z, B_{01}, B_{02}, B_{03}\}$  has to be determined from Zeeman spectroscopy measurements. The theoretical value of the zero-field frequency is expressed

as the sum  $f_{th,i} = f_{rot,i} + f_{hf,i}$  of the rotational frequency and the hyperfine frequency. A nonlinear least-squares adjustment of  $N$  Zeeman subcomponents can be exploited for absolute calibration of the experimental parameters  $\{V_k\}$  (at least  $N=9$  measurements are required). In order to estimate the uncertainties, the magnetic field dependence of eq. (4) is linearized around the design values of the parameters  $\{V_{k,0}\}$ :

$$\|\bar{B}(\{V_k\}_i; I_{1,i}, I_{2,i}, I_{3,i})\| = \|\bar{B}(\{V_{k,0}\}_i; I_{1,i}, I_{2,i}, I_{3,i})\| + \sum_k \frac{\partial \|\bar{B}(\{V_k\}_i; I_{1,i}, I_{2,i}, I_{3,i})\|}{\partial V_k} \Big|_{\{V_k\}=\{V_{k,0}\}} (V_k - V_{k,0}) \quad (5)$$

The  $9 \times 9$  covariance matrix  $G$  of the adjusted parameters may be estimated in function of the  $N \times N$  covariance matrix  $X$  of the input data  $b_i = (f_{exp,i} - f_{th,i})/\eta_i$ , and the  $N \times 9$

Jacobian matrix  $A = \left\{ \left( \frac{\partial \|\bar{B}_i\|}{\partial V_k} \right)_{i,k} \Big|_{\{V_k\}=\{V_{k,0}\}} \right\}$ :

$$G = (A^T X^{-1} A)^{-1} \quad (6)$$

The uncertainties of the input data are estimated as the quadratic sum of the contributions from the experimental uncertainty of the Zeeman subcomponent  $\delta f_{exp,i} = 71 \text{ mHz}$ , and from the theoretical uncertainties of the rotational frequency  $\delta f_{rot,i} = 10^{-12} \times f_{rot,i}$ , the hyperfine frequency  $\delta f_{hf,i} = 0.5 \text{ kHz}$ , and the Zeeman shift coefficient  $\delta \eta_i = 5 \text{ kHz/T}$ :

$$\delta b_i = \sqrt{\left( \frac{\delta f_{exp,i}}{\eta_i} \right)^2 + \left( \frac{\delta f_{rot,i}}{\eta_i} \right)^2 + \left( \frac{\delta f_{hf,i}}{\eta_i} \right)^2 + \left( \frac{\delta \eta_i (f_{rot,i} + f_{hf,i})}{\eta_i^2} \right)^2} \quad (7)$$

The nondiagonal elements of the  $X = \{x_{ij}\}$  matrix arise from the covariances between the theoretical parameters, expressed as:

$$x_{ij} = \left( \frac{\delta f_{exp,i}}{\eta_i} \right)^2 \times \delta_{ij} + \frac{\delta f_{rot,i} \delta f_{rot,j}}{\eta_i \eta_j} + \frac{\delta f_{hf,i} \delta f_{hf,j}}{\eta_i \eta_j} + \left( \frac{\delta \eta_i (f_{rot,i} + f_{hf,i})}{\eta_i^2} \right) \left( \frac{\delta \eta_j (f_{rot,j} + f_{hf,j})}{\eta_j^2} \right) \quad (8)$$

Let's consider an experiment designed with the following values of the parameters:  $k_1 = k_2 = k_3 = -10^{-4} \text{ T/A}$ ,  $\alpha_x = \alpha_y = \alpha_z = 0.05 \text{ rad}$ . and  $B_{01} = 3.82 \times 10^{-7} \text{ T}$ ,  $B_{02} = 2.09 \times 10^{-5} \text{ T}$ ,  $B_{03} = 4.34 \times 10^{-5} \text{ T}$ , respectively. The calibration is performed by Zeeman spectroscopy of the  $|v,L,F,S,J,J_z\rangle = |0,0,1,2,2,-2\rangle \rightarrow |0,2,1,2,4,0\rangle$   $\text{HD}^+$  transition. The Zeeman two-photon frequencies are measured for 27 sets  $(I_1, I_2, I_3) = (I_{off1} + n_1 I_0, I_{off2} + n_2 I_0, I_{off3} + n_3 I_0)$  of current intensities, where  $I_0 = 1 \text{ A}$ ,  $I_{off1} = 3.82 \text{ mA}$ ,  $I_{off2} = 208$

mA,  $I_{off3}=422$  mA, and  $n_{1,2,3}=0,1,2$ . The uncertainties of the values of the experimental parameters, that can be determined by the nonlinear adjustment, are estimated with eq. (4-8) :

$$\begin{aligned} \delta k_1 &= 5.4 \times 10^{-12} \text{ T/A}, \quad \delta k_2 = 1.6 \times 10^{-11} \text{ T/A}, \\ \delta k_3 &= 3.0 \times 10^{-11} \text{ T/A}, \quad \delta \alpha_x = 1.6 \times 10^{-7} \text{ rad}, \\ \delta \alpha_y &= 1.1 \times 10^{-7} \text{ rad}, \quad \delta \alpha_z = 7.9 \times 10^{-8} \text{ rad}, \\ \delta B_{01} &= 1.3 \times 10^{-11} \text{ T}, \quad \delta B_{02} = 1.2 \times 10^{-11} \text{ T}, \\ \delta B_{03} &= 1.5 \times 10^{-11} \text{ T}, \end{aligned} \quad (9)$$

Cancellation of the Earth's magnetic field in the ion trap is realized using the values of the current intensities that minimize eq. (4) :  $I_{null1} = -6.34$  mA,  $I_{null2} = 230$  mA,  $I_{null3} = 434$  mA. The uncertainty in setting the magnitude of the magnetic field is calculated with the root sum of squares of the contributions from the uncertainties of setting the current intensities (assumed at a fractional value of  $10^{-3}$  of each current intensity), and from the uncertainties of the experimental parameters determined previously by Zeeman spectroscopy. Particularly, the uncertainty in setting the null magnetic field in the ion trap is estimated at  $3.9 \times 10^{-8}$  T, if the theoretical uncertainties are neglected. In addition, a conservative estimation of the orientation uncertainty for a magnetic field with a given magnitude may be derived by assuming that the total magnetic field uncertainty is perpendicular to the magnetic field direction. If the three pairs of coils are driven by (-6.33 mA, 240 mA, 444 mA) currents, the magnitude of the magnetic field is  $1.7 \times 10^{-6}$  T and the orientation uncertainty is 30 mrad.

#### IV. CONCLUSION

This contribution describes the experimental setup and the calibration technique for the magnitude and the orientation of the magnetic field in an ion trap using precision Zeeman spectroscopy of  $\text{HD}^+$  ions. The Zeeman shift for a selected hyperfine component of a two-photon rotational transition is exploited for the determination of the

magnitude of a static magnetic field with an uncertainty at the  $10^{-11}$  T level. The approach allows coil calibration that enables to predict the magnitude and orientation of the magnetic field in function of the coil currents. The approach allows cancellation of the Earth's magnetic field at the 39 nT level. The orientation uncertainty of a magnetic field of 1.7  $\mu\text{T}$  is better than 30 mrad.

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