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# Characterization of a THz Electric Field using Precision Spectroscopy of Molecular Ions

Florin Lucian Constantin

Laboratoire PhLAM, CNRS UMR 8523, 59655 Villeneuve d'Ascq, France

FL.Constantin@univ-lille1.fr

**Abstract:** SI-traceable calibration of the amplitudes and phases of the Cartesian components of a THz electric field may be performed by comparing measurements of the lightshifts of  $\text{HD}^+$  two-photon rovibrational transitions with molecular ion theory predictions.

## 1. Introduction

The atomic spectroscopy was exploited to provide SI-traceable measurements, sub-wavelength resolution, accurate, stable and long-term reproducible detection of microwave electric fields [1]. The spectroscopy of the Rydberg states of the alkali metals was exploited to detect weak electric fields at the  $\mu\text{V}/\text{cm}$  level with a sensitivity limited by the photon shot noise [2]. The precision of the measurements of the  $\text{HD}^+$  transitions was significantly improved recently, reaching a fractional uncertainty of  $1.3 \times 10^{-11}$  for the fundamental rotational transition [3], and  $2.9 \times 10^{-12}$  for a two-photon rovibrational transition [4]. The ab-initio calculations provided accurate predictions for the  $\text{HD}^+$  energy levels [5,6] and their shifts in external fields [7,8]. The two-photon spectroscopy of  $\text{HD}^+$  has the potential to improve the fractional resolution and accuracy at the  $10^{-12}$  level or better [9]. This contribution proposes to exploit the sensitivity of a two-photon rovibrational transition of  $\text{HD}^+$  to external fields in order to detect a THz-wave that is off-resonantly coupled to the  $\text{HD}^+$  energy levels. The amplitude of the THz electric field may be calibrated subsequently by comparing the measurement of the induced frequency shift and the prediction of the molecular ion theory. A set of independent lightshift measurements can be converted into the amplitudes and the phases of the Cartesian components of the electric field of the THz-wave.

## 2. Results

The measurements may be performed with the experimental setups with  $\text{HD}^+$  ions trapped and sympathetically cooled with laser-cooled  $\text{Be}^+$  ions [3,4]. The  $\text{HD}^+$  transitions addressed in the sensing scheme are indicated in Fig. 1.(a). The measurements address the  $(v,L,F,S,J,J_z)=(0,0,1,2,2,2)\rightarrow(2,0,1,2,2,2)$  Zeeman component of the two-photon transition of  $\text{HD}^+$  at 55.909 THz (the energy levels are labelled with the quantum numbers for the  $\text{HD}^+$  spin coupling scheme [6]). The detection of this transition may be performed by dissociation of the  $(v,L)=(2,0)$  level with a 175 nm laser [9]. The experimental uncertainty of the frequency of the two-photon transition is estimated at 2.49 Hz, using the quantum projection noise limit in a single-ion spectroscopy experiment. A THz-wave slightly detuned to the Zeeman subcomponents of the  $(v,L)=(0,0)\rightarrow(0,1)$  rotational transition at 1.315 THz, or  $(v,L)=(2,0)\rightarrow(2,1)$  at 1.197 THz, may be detected by measuring the lightshift induced on the two-photon transition.

A static magnetic field is applied on the ion trap using three coil pairs oriented orthogonally along the axes of the Cartesian laboratory frame. The magnetic field is exploited to define the quantisation axis and to tune the  $\text{HD}^+$  energy levels. The magnetic field magnitude may be calibrated by comparing measurements by Zeeman spectroscopy of the  $(v,L,F,S,J,J_z)=(0,0,1,2,2,-2)\rightarrow(2,2,1,2,4,0)$  two-photon transition of  $\text{HD}^+$  with the predictions of the ab-initio theory. The uncertainty for magnetic fields smaller than  $10^{-4}$  T is estimated at the  $4 \times 10^{-10}$  T level, limited by the experimental uncertainty of the frequency of the two-photon transition.

The AC-Stark shifts induced by a THz-wave are quantified with the dynamic polarizabilities of the  $\text{HD}^+$  energy levels. They are calculated with their uncertainties using the second-order perturbation theory in function of a set of theoretical parameters (the  $\text{HD}^+$  energy levels, the rotational transition dipole moments, the radiative linewidths of the  $\text{HD}^+$  energy levels) and experimental parameters (the magnitude of the magnetic field, the polarization and the frequency of the THz-wave). The polarizability of a Zeeman level displays narrow dispersion-like resonances where the uncertainty increases at the frequencies of the electric dipole-allowed couplings to others levels (Fig. 1.(b)).

The performances of sensitivity and precision of the THz electric field measurements are evaluated by translating the uncertainties from the frequency measurements and from the ab-initio calculations of the theoretical parameters. For example, the measurement of the lightshift of the  $(v,L,F,S,J,J_z)=(0,0,1,2,2,2) \rightarrow (2,0,1,2,2,2)$  transition may be exploited to detect an electric field as low as  $6 \mu\text{V/m}$  of a THz-wave at  $1,314,947,502.3 \text{ kHz}$  with  $\pi$  polarization, by optimizing the value of the magnetic field in the ion trap. This result corresponds to a thirty times improvement comparing to the value of the weakest microwave electric field detected with the Rydberg atom spectroscopy method [2]. In addition, the precision of the calibration of the THz-wave with an intensity of  $1 \text{ W/m}^2$  based on lightshift measurements is at the  $10^{-3}$  level and is limited by the accuracy of the dynamic polarizability calculation.

The polarization ellipse (PE) of a THz electric field in three dimensions, described in the Cartesian laboratory frame with three real positive amplitudes and two phases  $(E_x, E_y, E_z, \phi_x, \phi_y)$ , may be fully characterized using lightshift measurements. Let's consider a reference THz-wave with a frequency of  $1,314,947,502.3 \text{ kHz}$ , an intensity of  $1 \text{ W/m}^2$ ,  $\sigma^+$  polarization, and propagating along a direction defined with Euler angles  $(\alpha, \beta) = (\pi/4, \pi/4)$  in the laboratory frame. The PE of the THz-wave may be determined from a set of six independent lightshift measurements for the  $(v,L,F,S,J,J_z) = (0,0,1,2,2,2) \rightarrow (2,0,1,2,2,2)$  transition for three magnetic fields  $B_1 = 10^{-6} \text{ T}$ ,  $B_2 = 5 \times 10^{-6} \text{ T}$ ,  $B_3 = 10^{-5} \text{ T}$  and two orientations defined with the Euler angles  $(\alpha, \beta) = (0,0)$  and  $(\alpha, \beta) = (0, \pi/2)$ . The lightshifts are simulated by adding to the theoretical values for the reference THz-wave a Gaussian noise with zero mean frequency and standard deviation given by the assumed uncertainty of the  $\text{HD}^+$  frequency measurements. The squared standard components of the THz electric field with respect to the two orientations of the quantization axis are subsequently derived using the differential dynamic polarizabilities of the two-photon transitions, evaluated as the theoretical value plus a random Gaussian noise with zero mean and standard deviation given by the calculation uncertainty. Then, the parameters of the THz electric field in the laboratory frame may be determined analytically from the calculated values of the squared standard components. The polarization ellipses of the THz electric field in the laboratory frame retrieved from 10 sets of lightshift measurements are represented in Fig. 1.(c). The uncertainties are at the  $(\delta E_x = \delta E_y = 1 \text{ mV/m}, \delta E_z = 10 \mu\text{V/m}, \delta \phi_x = 0.1 \text{ rad}, \delta \phi_y = 0.1 \text{ rad})$  levels.

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## Figure caption

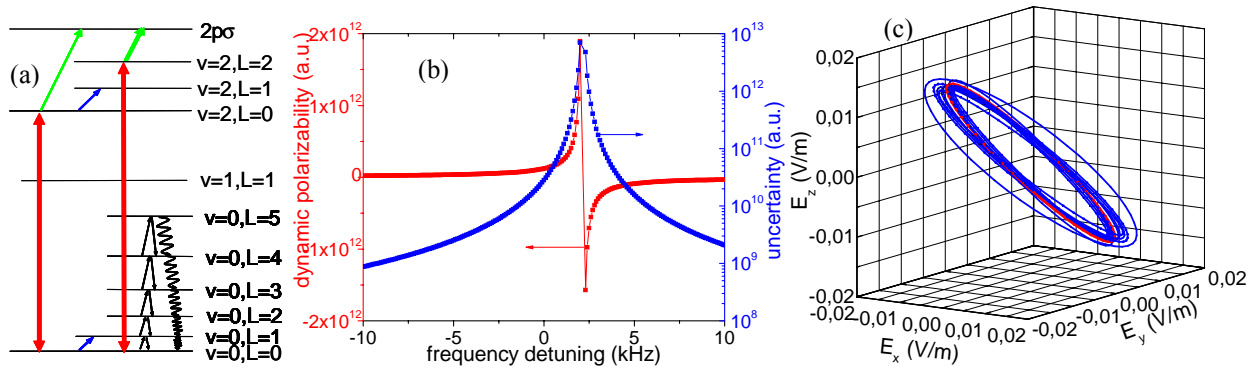


Fig. 1. (a)  $\text{HD}^+$  transitions for THz-wave and magnetic field sensing : two photon infrared transitions (red lines), dissociation (green lines), blackbody radiation driven transitions (black lines), THz-wave driven dipole couplings (blue lines). (b) Frequency dependence of a term in the dynamic polarizability of the  $(v, L, F, S, J, J_2) = (0, 0, 1, 2, 2, 2)$  level (red curve, left axis) and its uncertainty (blue curve, right-axis). This term characterizes in the lightshift induced by a  $\sigma^+$ -polarized THz-wave. Detuning to 1314945902.3 kHz. Magnetic field  $10^{-4}$  T. (c) The reference polarization ellipse of the THz-wave (red curve). The polarization ellipses determined from lightshift measurements (blue curve).