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Two-Photon Rotational Spectroscopy of Cold Trapped HD⁺ Ions for Improved Characterization of THz Electric Fields

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Abstract: Comparison of two-photon rotational spectroscopy measurements of cold trapped HD⁺ ions with ab-initio quantum theory predictions may improve significantly the characterization of the amplitudes and phases of the Cartesian components of a THz electric field.

1. Introduction

A Doppler-free rotational spectroscopy approach applied to the hydrogen molecular ions enabled improved accuracy and record-high resolution [1]. The molecular theory allowed to predict their rovibrational energy levels with fractional accuracy beyond the 10^{-11} level [2], their hyperfine energy levels with 10^{-6} level precision [3], and their energy level shifts in external fields [4,5]. The comparison between the theoretical and the experimental frequencies was exploited for a test of the quantum electrodynamics, to search for a deviation of the interaction between nuclei from the Coulomb force and for improving the determination of a set of fundamental constants. The comparison experiment-theory was extended to the frequency shifts induced on HD^+ transitions by external fields in order to calibrate them against the values of the fundamental constants and the SI second [6,7]. Double-resonance two-photon spectroscopy has potential to increase the fractional resolution and accuracy beyond the 10^{-12} level [8]. This contribution addresses an improved THz sensing technique that exploits the lightshifts induced on a two-photon rotational transition by a THz-wave that is coupled off-resonantly to the intermediate rotational level.

2. Results

The approach is based on the experimental setups developed for Doppler-free spectroscopy of HD^+ [1,9,10]. A double-resonance two-photon spectroscopy scheme is exploited to probe the hyperfine components of the $(v,L)=(0,0)\rightarrow(0,2)$ transition of HD^+ [8]. The uncertainties of the frequency measurements are estimated with the limit for the molecular ion quantum projection noise at ~ 71 mHz. A static magnetic field is applied to the ion trap with three pairs of coils having an ideal, orthogonal orientation that defines the Cartesian laboratory reference frame. The strength of the magnetic field is calibrated by comparing Zeeman spectroscopy measurements of the two-photon rotational transition $(v,L,F,S,J,J_z)=(0,0,1,2,2,-2)\rightarrow(0,2,1,2,4,0)$ at 1.968 THz with the predictions of the ab-initio theory. The sensitivity of this method is limited by the frequency measurement uncertainty at 1.4×10^{-11} T. A magnetic field smaller than 10^{-4} T may be calibrated using the comparison theory-experiment with an accuracy better than 7×10^{-11} T.

A THz-wave slightly detuned to the $(v,L,F,S,J,J_z)=(0,0,1,2,2,2)\rightarrow(0,1,1,2,3,3)$ transition at 1.315 THz is detected using the lightshifts induced on the two-photon rotational transition $(v,L,F,S,J,J_z)=(0,0,1,2,2,2)\rightarrow(0,2,1,2,4,4)$. The energy level shifts are quantified in function of

the squared standard components of the THz electric field relative to the quantization axis and of the dynamic polarizabilities of HD^+ , calculated here with their uncertainties using a set of theoretical parameters and the experimental magnetic field strength. Using the rotational transitions with narrow radiative linewidths reduces the uncertainty in magnetic field and lightshift measurements and enables an improved THz sensing method comparing to the approach based on rovibrational transitions of HD^+ [7]. The uncertainty of the measurement of a THz electric field with a given intensity, frequency and polarization is estimated by translating uncertainties from lightshift measurements and from calculations of dynamic polarizabilities. The method enables weak THz electric field detection with a limit estimated at $7 \mu\text{V/m}$ for a THz-wave at $1,314,947,502.3 \text{ kHz}$ with σ^- polarization using an optimal magnetic field strength. That is a significant improvement comparing to the value of the weakest microwave electric field detected by Rydberg spectroscopy of alkali vapors method [11]. In addition, for each lightshift measurement the value of the magnetic field is optimized in order to minimize the uncertainty of the THz-wave intensity. The performances of this THz electrometry method based on the two-photon rotational transition of cold trapped HD^+ are displayed in Fig. 1.(a).

A THz electric field, 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 from the lightshifts of the two-photon rotational transition. This contribution addresses the case of a THz-wave with a frequency of $1,314,947,502.3 \text{ kHz}$, an intensity of 1 W/m^2 , and a linear polarization oriented towards a direction defined with Euler angles (θ, ϕ) in the laboratory frame. The parameters of the THz-wave may be determined from a set of six independent lightshift measurements for three magnetic field strengths $B_1=1 \mu\text{T}$, $B_2=5 \mu\text{T}$, $B_3=10 \mu\text{T}$ and two orientations defined with Euler angles $(\alpha, \beta)=(0,0)$ and $(\alpha, \beta)=(0, \pi/2)$. The experimental lightshifts are calculated by shifting the theory predictions with random values with a Gaussian distribution with zero mean and standard deviation given by the lightshift measurement uncertainty. The dynamic polarizabilities are estimated by adding to the theoretical value a random error with a Gaussian distribution with zero mean and standard deviation given by the polarizability calculation uncertainty. That allows to derive analytically six squared standard components of the THz electric field and, by inversion, the parameters of the THz electric field in the laboratory frame. This procedure is exploited to estimate the uncertainties of the measurements of THz-waves with arbitrary polarization (Fig. 1(b.1-b.5)) using 10 sets of lightshift measurements. For a broad range of polarization orientations, the uncertainties for

E_x , E_y are less than $5 \mu\text{V/m}$, for E_z less than $0.3 \mu\text{V/m}$, and for ϕ_x , ϕ_y less than 5 mrad , that corresponds to an improvement by three orders of magnitude comparing to the previous approach based on rovibrational transitions [7].

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Figure captions

Fig. 1. (a) Combined experimental and theory errors for the THz-wave intensity calibration (red line) and the limit imposed by the frequency measurement error (blue line). (b) Uncertainties of the amplitudes (b.1-b.3) and the phases (b.4, b.5) of the Cartesian components of the electric field of a linearly polarized THz-wave derived from lightshift measurements in function of the orientation of the polarization vector.

