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Precision Spectroscopy of Cold Trapped HD^+ Ions for Characterization of the Polarization Ellipse of a THz-Wave

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Abstract: Comparison of accurate ab-initio molecular theory calculations and precise measurements of the lighshifts of the two-photon rovibrational transitions of HD^+ may provide SI-traceable calibration of the electric field magnitude and polarization state of a THz-wave.

I. INTRODUCTION

Atomic spectroscopy can provide SI-traceable measurements, sub-wavelength resolution, accurate, stable and long-term reproducible detection of microwave and sub-THz electric fields [1]. The Rydberg states of alkali metals were exploited to build compact sensors based on vapour cells that allowed weak electric field detection at the $\mu\text{V}/\text{cm}$ level with a sensitivity limited by the photon shot noise [2]. The metrology of the THz electric fields is addressed here with precision measurements with cold trapped hydrogen molecular ions (HMI). The rovibrational transitions of HD^+ were measured with 10^{-9} precision spectroscopy [3]. Doppler-free spectroscopy at 1.3 THz of the fundamental rotational transition of HD^+ ions in the Lamb-Dicke regime improved the precision at the 10^{-10} level [4]. Two-photon THz/IR spectroscopy have the potential to improve the fractional resolution and accuracy at the 10^{-12} level or better [5]. HMI benefit from accurate ab-initio calculations for their energy levels [6] and of the frequency shifts induced by interaction with external fields [7,8]. This contribution proposes a new method to calibrate the electric field of a THz-wave that is off-resonantly coupled to the HD^+ energy levels by exploiting the comparison of the measurements of the lightshift induced on a two-photon rovibrational transition with the predictions of the molecular ion theory. A set of independent lightshift measurements can be converted into the amplitudes and the phases of the components of the THz electric field in the Cartesian laboratory frame.

II. RESULTS

The precision measurements proposed here can be performed with the experimental setups with HD^+ ions trapped and sympathetically cooled with laser-cooled Be^+ ions, by using a detection based on resonant enhanced multiphoton dissociation (REMPD) [3,4]. The HD^+ energy levels addressed in the THz sensing scheme are depicted in Fig. 1(a). Here, the measurements address a Zeeman subcomponent of the $(v,L)=(0,0)\rightarrow(2,0)$ two-photon transition of HD^+ at 55.909 THz. The detection is performed by dissociation of the $(v,L)=(2,0)$ level with a 175 nm laser [5]. A set of rate equations is developed to describe the time dependence of the populations of the HD^+ energy levels during the spectroscopy scheme, by taking into account the interaction with blackbody radiation at room temperature. The lineshape of the two-photon resonance (Fig. 1.(b1)) displays a full-width at half maximum linewidth of 20 Hz, for a two-photon transition rate of 10 s^{-1} , a dissociation rate of 200 s^{-1} , and a REMPD time of 10 s. The limit of the frequency uncertainty due to the molecular ion quantum projection noise is estimated at 2.49 Hz. The THz-wave, slightly detuned to the Zeeman subcomponents of the rotational transition $(v,L)=(0,0)\rightarrow(0,1)$ at 1.315 THz or $(v,L)=(2,0)\rightarrow(2,1)$ at 1.197 THz, is detected via the lightshift induced on the two-photon transition.

A static magnetic field, applied on the ion trap using three coil pairs oriented orthogonally along the axes

of the laboratory frame, is exploited to tune the HD^+ energy levels and to define the quantisation axis. The direction and magnitude of the magnetic field are calibrated using 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 (the magnetic levels are labelled with the quantum numbers for the HD^+ spin coupling scheme [8]). The calibration accuracy for a 10^{-4} T magnetic field is estimated at the 10^{-7} T level, which is limited by the accuracy of the theoretical predictions.

The AC-Stark shifts are quantified with the dynamic polarizabilities of the HD^+ energy levels. They are calculated here with their uncertainties, using the second-order perturbation theory, in function of a set of theoretical parameters (the energy levels of HD^+ , the rotational transition dipole moments, the radiative linewidths of the 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 magnetic state displays narrow dispersion-like resonances where the uncertainty increases, at the frequencies of the electric dipole-allowed couplings to others energy levels (Fig. 1(b2)).

The performances of precision and sensitivity of the THz electric field measurements are evaluated by assessing the impact of the uncertainties from the frequency measurements and from the ab-initio calculations of the theoretical parameters. For a THz-wave σ^- polarized and detuned by 1.6 MHz to the $(v,L,F,S,J) = (0,0,1,2,2)\rightarrow(0,1,1,2,3)$ hyperfine transition, the lowest THz electric field that can be detected is 11 $\mu\text{V/m}$. A THz electric field with the same frequency and polarisation and with an amplitude of 10 mV/m can be calibrated with a fractional accuracy of 5×10^{-3} , limited by the uncertainties of the theoretical parameters.

The polarization ellipse (PE) of the THz electric field is defined in the laboratory frame with three real positive amplitudes and two phases $(E_x, E_y, E_z, \phi_x, \phi_y)$. This contribution proposes an algorithm to determine the PE from a set of five independent lightshift measurements for two different orientations of the magnetic field, specified with the Euler angles (α, β) in the laboratory frame. For example, a reference THz-wave at 1314947502.3 kHz with $(E_x = E_y = E_z = 15.83 \text{ mV/m}, \phi_x = \pi/4 \text{ rad}, \phi_y = \pi/3 \text{ rad})$ may be completely characterized with the measurements of the lightshifts for the following magnetic fields $B_1 = 10^{-6}$ T, $B_2 = 5 \times 10^{-6}$ T, $B_3 = 10^{-5}$ T with orientations defined with the Euler angles $(\alpha, \beta) = (0, 0)$ and $(\alpha, \beta) = (0, \pi/2)$.

Precisely, the lightshifts for the $(v,L,F,S,J,J_z) = (0,0,1,2,2,2)\rightarrow(2,0,1,2,2,2)$ transition are simulated for the reference THz-wave by adding to the theoretical values a Gaussian noise with zero mean frequency and standard deviation, given by the molecular ion quantum projection noise in frequency measurements. The standard components of the THz-wave are subsequently recalculated with the relevant set of dynamic polarizabilities, evaluated as the theoretical value plus a random Gaussian noise with zero mean and standard deviation given by the calculation uncertainty. The retrieved polarization ellipse of the THz

electric field in the laboratory frame is represented in Fig. 1(c) for 10 comparisons theory-experiment using the simulated lightshifts. The uncertainties of the amplitudes and phases of the THz electric field in the laboratory frame at the ($\delta E_x = \delta E_y = 1$ mV/m, $\delta E_z = 1$ μ V/m, $\delta\phi_x = 0.1$ rad, $\delta\phi_y = 10$ mrad) levels.

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Figure captions

Fig. 1. (a) HD^+ energy levels for the THz sensing scheme. Two photon transitions (red lines), dissociation (green lines), BBR-driven transitions (black lines), THz-wave driven dipole couplings (blue lines).

(b1) Calculated lineshape of the two-photon rovibrational resonance. (b2) Frequency dependence of a term in the dynamic polarizability of the $(v,L,F,S,J,J_z) = (0,0,1,2,2,2)$ level (left axis) and its uncertainty (right-axis). This term characterizes in the lightshift induced by a σ^+ -polarized THz-wave. Detuning to 1314945902.3 kHz. Magnetic field 10^{-4} T.

(c) The polarization ellipse of the THz-wave (blue line). Polarization ellipses determined from lightshift measurements (red lines).

