New bounds on Lorentz violation from a composite pulse method in a trapped ion

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Gravity is not understood at a quantum mechanical level. In an attempt to formulate a single quantum-consistent theory of the four known fundamental forces, it is suggested that spontaneous breaking of Lorentz symmetry might occur at the Planck scale¹,². In the low-energy limit, such a Lorentz violation would give rise to small shifts of energy levels with non-spherical atomic orbitals³. These could be observed with precision spectroscopy of atoms in a Michelson-Morley type experiment using Earth’s rotation⁴–⁶. In this work we perform such an experiment in the long-lived electronic ²F7/2 manifold of the Yb⁺ ion. We exploit the high intrinsic susceptibility of this state to Lorentz violation⁷ and apply an elaborate radio-frequency spin-echoed Ramsey sequence⁸ to investigate the isotropy of space-time with a single trapped ion. A robust composite pulse sequence allows us to extend the coherence time to more than 1 s and accurately compare the orthogonally oriented sublevels of the Zeeman manifold. With a three times higher sensitivity to Lorentz violation compared to the best test to date⁶, we improve the constraints on all the measured Lorentz symmetry breaking coefficients and set their bounds at the 10⁻²¹ level. These results represent the most stringent test of this type of Lorentz violation in the electron-photon sector. The method is readily extendable to multiple ions in Coulomb crystals, enabling improved tests of Lorentz symmetry in the near future.

The standard model (SM) of particle physics describes non-gravitational interactions between all particles and fields, while gravitation is described by general relativity in a classical manner. Together, they have explained many physical phenomena observed in the universe remarkably well, but an accurate description of gravity at the quantum level
is lacking. A number of theories that attempt to unify the SM and gravitation at the Planck scale suggest that spontaneous breaking of Lorentz symmetry might occur\textsuperscript{1,9–11}. Lorentz symmetry states that the outcome of a local experiment does not depend on the orientation or the velocity of the apparatus\textsuperscript{12}. A suppressed effect emerging from Lorentz violation (LV) at the Planck scale could be observed with low-energy experiments in the laboratory. Accurate spectroscopic measurements in trapped particles have reached fractional uncertainties beyond the natural suppression factor\textsuperscript{13}, which makes a hypothetical LV measurable in such systems. Furthermore, at high energies, LV could be suppressed by super-symmetry\textsuperscript{14}. Therefore, accurate low-energy measurements in atoms are suitable to search for LV and complement existing bounds set at high energies with, e.g., particle colliders and astrophysical observations\textsuperscript{15–17}.

Laboratory tests of Lorentz symmetry are based on a similar principle as introduced by Michelson and Morley, who used a rotating interferometer to measure the isotropy of the speed of light\textsuperscript{18}. Improved bounds on LV for photons have been realized by a variety of experiments involving high-finesse optical and microwave cavities, see e.g. refs. 12, 19–22. Spectroscopic bounds for protons and neutrons were set first using nuclear magnetic resonance on bare nuclei and later with co-magnetometers\textsuperscript{23–25}. More recently, the bounds on LV in the combined electron-photon sector have been explored using precision spectroscopy in trapped ions\textsuperscript{4–6}. These experiments compare energy levels with differently oriented, relativistic, non-spherical electron orbitals as the Earth rotates. Strong bounds on LV were set with trapped \textsuperscript{40}Ca\(^+\) ions, where a decoherence-free entangled state of two ions in the electronic $^2D_{5/2}$ manifold was created to suppress ambient noise\textsuperscript{4,5}. The relatively short 1.2 s radiative lifetime of the $^2D_{5/2}$ state in Ca\(^+\) and the requirement for high fidelity quantum gates limit the scalability and, ultimately, the sensitivity of LV tests with this scheme\textsuperscript{5}. The highly relativistic $^2F_{7/2}$ state in the Yb\(^+\) ion is an order of magnitude more sensitive to LV than the $^2D_{5/2}$ state in Ca\(^+\)\textsuperscript{7,8} and its radiative lifetime was measured to be about 1.6 years\textsuperscript{26}. These beneficial properties were recently exploited in a 45-day comparison of two separate state-of-the-art single-ion optical \textsuperscript{171}Yb\(^+\) clocks, both with a systematic uncertainty at the $10^{-18}$ level, resulting in a more than ten-fold improvement of the LV bounds\textsuperscript{6}.

In this work we present new bounds on LV in the electron-photon sector using a simple, robust and scalable measurement scheme in a single trapped ion experiment. We combine the high susceptibility of the $^2F_{7/2}$ state to LV with an improved radio-frequency (rf) spin-echoed Ramsey sequence\textsuperscript{8,27} to make a direct energy comparison between the nearly orthogonal atomic orbitals of the F-state Zeeman manifold within a single trapped \textsuperscript{172}Yb\(^+\) ion. In this manner, we fully exploit the most sensitive stretched $m = \pm 7/2$ states\textsuperscript{8} and eliminate the requirement of optical clock operation or high-fidelity quantum gates. With the applied composite rf pulse sequence, the influence of fluctuations in the ambient magnetic field on the coherence time is suppressed during the Ramsey measurement, extending the interrogation time $T_D > 1$ s. As a result, the sensitivity of the demonstrated method to LV, which scales as $\sigma_{LV} \propto \sqrt{1/T_D N_{\text{ion}}}$, is unprecedented already for a single ion ($N_{\text{ion}} = 1$). The rf sequence is robust against both temporal and spatial field inhomogeneities and can be readily applied to a string of $N_{\text{ion}}$ trapped ions to further increase the sensitivity to LV in the near future.

The constraints on LV extracted in this work are quantified in the theoretical framework of the standard model extension (SME)\textsuperscript{28}. The SME is an effective field theory in which the SM Lagrangian is extended with all possible terms that are not Lorentz invariant. It is a platform in which LV of all SM particles are described, enabling comparisons between experimental results from many different fields\textsuperscript{29}. In spectroscopic experiments,
a violation of Lorentz symmetry can be interpreted as LV of electrons or photons, because there is no preferred reference system. In this work, we interpret the results as a difference in isotropy between photons and electrons, similar as in refs. 4, 6.

LV in the combined electron-photon sector is quantified by adding a symmetry-breaking tensor \( c'_{\mu\nu} = c_{\mu\nu} + k_{\mu\nu}/2 \) to the SM Lagrangian\(^7,28\), where \( c_{\mu\nu} \) and \( k_{\mu\nu} \) describe LV for electrons and photons, respectively. For simplicity, the prime is omitted throughout the rest of this work and the extracted coefficients are those of the combined \( c'_{\mu\nu} \) tensor, which is taken as traceless and symmetric. The components of the \( c_{\mu\nu} \) tensor are frame dependent. A unique definition of the symmetry breaking tensor \( c_{MN} \) exists in the Sun-centered, celestial, equatorial frame (SCCEF), illustrated in Fig. 1. In order to make comparisons with other experiments, the \( c_{\mu\nu} \) tensor defined in our local laboratory frame is transformed to the SCCEF to constrain the components of the \( c_{MN} \) tensor. The full derivation of the transformation can be found in the Methods section.

In a bound electronic systems, LV leads to a small energy shift\(^7,8\)

\[
\delta H = -\frac{1}{6m_e} C^{(2)}_{0} T^{(2)}_0,
\]

where \( m_e \) is the electron mass, the \( T^{(2)}_0 = p^2 - 3p_z^2 \) operator depends on the direction of the electron’s momentum and \( C^{(2)}_{0} \) contains elements of the \( c_{MN} \) tensor. For a state with total angular momentum \( J \) and projection \( m \) onto the quantization axis \( \hat{z} \), the matrix element of the \( T^{(2)}_0 \) operator is given by\(^8\)

\[
\langle J, m | T^{(2)}_0 | J, m \rangle = \frac{-J(J+1) + 3m^2}{\sqrt{(2J+3)(J+1)(2J+1)J(2J-1)}} \times \langle J||T^{(2)}||J \rangle.
\]

Equations (1) and (2) show that in the SCCEF, LV manifests itself as an energy shift that modulates with Earth’s rotation. The magnitude of this shift is dependent on both \( m^2 \) and the reduced matrix element \( \langle J||T^{(2)}||J \rangle \). The value of the latter is particularly high for the \( ^2F_{7/2} \) manifold in the Yb\(^+\) ion\(^7,8\). The goal of this experiment is to test LV in a single trapped \(^{172}\)Yb\(^+\) ion by measuring the energy difference between \( m \)-substates in the \( ^2F_{7/2} \) manifold as the Earth rotates.

The experiment is performed with a single ion, stored in a linear rf Paul-trap, see Fig. 1b. It is cooled to the Doppler limit of around 0.5 mK on the dipole allowed transition near a wavelength of 370 nm, assisted by a repumper near 935 nm. A set of coils is used to define the quantization axis of \( B = 221 \mu \text{T} \), which lies in the horizontal plane with respect to Earth’s surface points 20\(^\circ\) south of east, see Fig. 1a. Active feedback is applied on auxiliary coils in three dimensions to stabilize the magnetic field. The \( ^2F_{7/2} \) state can be efficiently populated via coherent excitation of the highly-forbidden electric octupole (E3) transition (Fig. 1c) using an ultra-stable frequency-doubled laser at 934 nm\(^30\), which is either stabilized only to a cryogenic silicon cavity via a frequency comb\(^31\) or, optionally, to the single-ion optical \(^{171}\)Yb\(^+\) clock\(^32\). A Rabi frequency of \( \Omega_{\text{E3}}/2\pi = 10 \text{ Hz} \) is achieved on the E3 transition. More details on the experimental apparatus can be found in the Methods section.

The free evolution of a substate \( |^2F_{7/2}, m \rangle \) interacting with a magnetic field \( \mathbf{B} \equiv B_z \hat{\mathbf{z}} \), is given by the Hamiltonian \( \mathcal{H}_{\text{free}} = \mathcal{H}_{\text{lin}} + \mathcal{H}_{\text{quad}} = \mu B_z J_z + \kappa J_z^2 \), where \( \mu \) is the magnetic moment. The quadratic term in the Hamiltonian gives rise to an energy shift according to \( E_m/\hbar \equiv \kappa/2\pi \times m^2 \). The value of \( \kappa = \kappa_0 + \kappa_{\text{LV}} \) is dependent on the quadrupole shift for the trapped ion in the \( ^2F_{7/2} \) state and a possible shift due to LV, respectively. The
A schematic overview of the experimental principle. **a** In the laboratory frame (LF) coordinate system $z$ points vertically upwards, $y$ points North and $x$ points east. The fixed quantization axis $B$ (pink arrow) in the LF coordinate system lies in the horizontal plane and points $20^\circ$ south of $x$ and probes different directions in the SCCEF as the Earth rotates around its axis ($\omega_\oplus$) and orbits the Sun ($\Omega_\odot$). **b** In the LF a single $^{172}\text{Yb}^+$ ion is trapped in a segmented rf trap. Two coils generate a quantization field of $B = 221 \, \mu T$. The electron orbitals of the $m = \pm 1/2$ and $m = \pm 7/2$ Zeeman sublevels in the $^{2}F_{7/2}$ state orient themselves orthogonally to each other. The $^{2}F_{7/2}$ state is population via the E3 transition near 467 nm. The $m$ substates are coupled via the rf magnetic field created with a coil that is placed 5.5 cm above the ion. **c** A simplified energy level diagram of $^{172}\text{Yb}^+$, showing the optical E3 transition and the transitions near 370 nm and 935 nm used for Doppler cooling and state detection.

The contribution of LV to $\kappa$ is given by\(^8\)

\[
\frac{\kappa_{\text{LV}}}{2 \pi} = 5.1 \times 10^{15} \text{ Hz} \times C_{0}^{(2)},
\]

where $C_{0}^{(2)}$ contains components of the $c_{M,N}$ tensor in the SCCEF, see Methods.

A modulation of the quadratic contribution to the Zeeman splitting in the $^{2}F_{7/2}$ manifold is measured with rf Ramsey spectroscopy. The $m$-levels are coupled via a rf magnetic field supplied to the ion by a resonant LC circuit. The coupling term in the Hamiltonian is given by $H_{\text{coupling}} = \Omega_F \cos(\omega_{rf} t + \phi) J_x$, where $\Omega_F/2\pi = 33 \, \text{kHz}$ is the multilevel Rabi frequency and $\omega_{rf}$ and $\phi$ are the frequency and the phase of the rf field, respectively. The rf frequency is close to resonance with the, to first order, equidistant $m$-levels given by $\omega_{rf}/2\pi = \mu B_z / \hbar + \delta(t)/2\pi \approx 3.5 \, \text{MHz}$, where $\delta(t)$ is a small detuning from temporal drifts in the ambient magnetic field. The full Hamiltonian of the system in the interaction picture after applying the rotating wave approximation is given by

\[
H = \delta(t) J_z + \kappa J_z^2 + \Omega_F [J_x \cos(\phi) - J_y \sin(\phi)],
\]

where the changes in $\delta(t)$ and $\kappa$ should be much slower than the $\pi$-pulse time of $t_\pi = \pi/\Omega_F = 15 \, \mu s$.

A composite rf pulse sequence, based on a spin-echoed Ramsey scheme, is implemented to mitigate the influence of $\delta(t)$, while retaining a high sensitivity to variations of $\kappa$. A
A schematic overview of the full experimental sequence is shown in Fig. 2 b. Stable long-term operation of the experiment is required to resolve oscillation periods related to a potential LV, i.e. 11.967 and 23.934 hours. Especially the center frequency of the E3 transition is sensitive to external perturbations from, e.g., magnetic field drifts and intensity fluctuations. Therefore, a 4-point servo-sequence of two opposite Zeeman transitions is sensitive to variations of $\kappa_{{\rm q}}$, see e.g. ref. 34. On average a population transfer of 80% is realized to the $^2F_{7/2}$ state via $^2S_{1/2}$ center frequency. For details on this technique, see the Methods section. The corresponding optimal Ramsey dark time is $T_D = 1.15 \text{ s}$. A schematic overview of the full experimental sequence is shown in Fig. 2 b.
Fig. 2  Pulse sequences. a The applied composite rf pulse sequence is a more robust version of a spin-echoed Ramsey scheme. A Ramsey sequence of \( T_{\text{rf}} = 1236.27 \text{ ms} \) starts and ends with a \( \pi/2 \)-pulse. A \( 2 \times 2.15 \text{ ms} \)-long modulation sequence of \( 10 \pi \)-pulses with phases \( \phi_i \) (\( i = 1 \ldots 10 \)) each spaced by \( 2t_{\text{w}} = 200 \mu\text{s} \) is repeated \( N = 575 \) times to cancel dephasing due to ambient magnetic field noise. The phases \( \phi_i \) are chosen such (see text) that the sequence is robust against pulse errors. b Every \( n = 50 \) measurements, a 4-point servo sequence (E3 servo) of 40 s is applied to follow the E3 center frequency, after which the rf sequence is compiled for \( 2.5 \text{ s} \). The measurement sequence consists of two \( 50 \text{ ms} \)-long E3 pulses (E3), the rf pulse sequence (rf) of \( 1236.27 \text{ ms} \), and four detection pulses (D) of \( 2.5 \text{ ms} \) and a sequence at the end of \( 20 \text{ ms} \) for state preparation (R) for the next measurement run. During post-selection, the data points are considered valid if the ion was in the desired state during the first three detection pulses.

valid when the ion was in the correct state at both the second and the third detection stage. After post-selection of valid data points an average contrast of 0.77(6) is reached. Including additional overhead from compilation time, data points are obtained at a rate of \( 1/191 \text{ s}^{-1} \).

The data acquired over a period of 912 h, with an up-time of 591 h, is shown in Fig. 3 a. Data points are averaged in equidistant bins of 15 min. The measured population is decomposed in two parts \( P_1 = P(\kappa_q) + P(\kappa_{LV}) \), where \( P(\kappa_q) \) gives rise to a constant offset of \( P_0 = 0.39 \) and \( P(\kappa_{LV}) = dP_1/d\kappa \times \kappa_{LV} \) contains a potential LV signal. To extract \( \kappa_{LV} \) from \( P_1 \), a high-pass filter is applied with a cutoff frequency of \( \nu_c = 5 \mu\text{Hz} \), removing \( P_0 \) and slow variations (\( \tau_{\text{drift}} < 2.5 \text{ days} \)) caused by drifts in the E3 excitation probability and the slope \( dP_1/d\kappa|_{\kappa=0.13(3)} = -4.4(4) \) is calculated. More details on the data handling and the measurement sensitivity can be found in the Methods section.

In search of LV at Fourier frequencies of \( \omega_{\text{E3}} \) and \( 2\omega_{\text{E3}} \), the data is fitted globally to the fit function

\[
\kappa_{LV} = 2\pi \times 5.1 \times 10^{15} \times [-3 \sin(2\chi) c_{XZ} \cos(\omega_{\text{E3}}T) - 3 \sin(2\chi) c_{YZ} \sin(\omega_{\text{E3}}T) - \frac{3}{2} c_{X-Y} \sin^2(\chi) \cos(2\omega_{\text{E3}}T) - 3 c_{XY} \sin^2(\chi) \sin(2\omega_{\text{E3}}T)],
\]

from which the individual components of the \( c_{MN} \) tensor in the SCCEF are extracted, where \( c_{X-Y} = c_{XX} - c_{YY} \). The fit overlays the data in Fig. 3 a and the residuals from the fit are shown in Fig. 3 b, from which the reduced chi-square of \( \chi^2 = 0.92 \) is extracted. The fitted values of the components of the \( c_{MN} \) tensor are given in Tab. 1. For comparison, the values obtained by refs. 5, 6 are also presented. The uncorrelated linear combination of the fit parameters, calculated by diagonalizing the covariance matrix, are given in the lower part of Tab. 1. The spectral content of the data, shown in Fig. 3 c, is analyzed...
using a Lomb-Scargl periodogram\textsuperscript{35}, which is specifically suited for spectral analysis of irregularly spaced data. The statistical significance level, i.e. p-values\textsuperscript{35}, of 50\% and 95\% are indicated by horizontal lines.

The fit results show that the extracted values for $c_{X-Y}$, $c_{XZ}$ and $c_{YZ}$ are consistent with zero within a 1\sigma uncertainty. Only $c_{XY}$ shows a 1.1\sigma deviation from zero, but spectral analysis does not show a significant Fourier component at 2\omega_{⊕}. Therefore, we conclude that we do not find evidence of LV, in agreement with previous work\textsuperscript{5,6}. The stability of the data points is analyzed by calculating the Allan deviation, as shown in Fig. 3d. The data averages down as $\sigma_\kappa = 372(9)\text{ mrad }\tau^{-1/2}$, which is 29(3)\% higher than what is expected from quantum projection noise.

The presented results set the most stringent bounds on this type of LV in the combined electron-photon sector. The uncertainties on all the extracted coefficients of the $c_{MN}$ tensor are at the $10^{-21}$ level. Due to the experimental geometry, see Methods, a higher sensitivity is reached for signals that oscillate at 2\omega_{⊕} than those that oscillate at \omega_{⊕}. The bounds on $c_{X-Y}$ are improved by a factor of 2.2 in 1.8 times less total averaging time, demonstrating a 3-times higher sensitivity to LV compared to the previous most sensitive measurement\textsuperscript{6}. The tightest constraint of $3.9 \times 10^{-21}$ is achieved on $c_{XY}$.

In this work, coefficients of the first and second harmonic order of the sidereal day modulation frequency were considered. However, due to the high total angular momentum of the $^2F_{7/2}$ state, the applied method is sensitive to LV at harmonics of up to sixth order\textsuperscript{36,37}. Therefore, in combination with new many-body calculations, experimental constraints can be translated into bounds on a larger number of coefficients in the future\textsuperscript{37}.

The demonstrated method is technically less demanding and more robust than alternative methods requiring optical clock operation or high-fidelity entanglement gates\textsuperscript{5,6}. It
is readily scalable to multiple $N_{\text{ions}}$ in linear ion Coulomb crystals to further enhance the sensitivity by a factor $\sqrt{N_{\text{ion}}}$. The implemented composite rf pulse sequence is highly robust against errors originating from spatial and temporal fluctuations in both the ambient field and the rf field. Therefore, the coherence time is not expected to significantly decrease for larger ion numbers, in contrast to measurement schemes that rely on entangled states. For efficient population transfer via the E3 transition, advanced cooling techniques, such as EIT or Sisyphus cooling, might be advantageous in larger ion crystals. The long lifetime of the $^2F_{7/2}$ state in Yb$^{+26}$ does not significantly limit the coherence time and with several technical improvements, longer interrogation times might be reached. Therefore, the boundaries of Lorentz symmetry tests can be pushed to the $10^{-22}$ level with a string of 10 ions in the near future with the presented method.
Methods

Experimental details

A single $^{172}$Yb$^+$ ion is trapped in a segmented rf Paul-trap$^{41,42}$. The radial confinement is set with an rf electric field supplied by a resonant circuit at a frequency of $\Omega_{rf}/2\pi = 24.38$ MHz, while the axial confinement is set by dc voltages supplied to the trapping segment and the neighboring segments. With the applied confinement, the secular frequencies are $\omega$(rad1, rad2, ax)/$2\pi = (775, 510, 266)$ kHz. Micromotion is measured on a daily basis with the photon correlation technique$^{43}$ and compensated in three dimensions to typically $E_{rf} < 100$ V/m. The quantization field of $B = 221$ $\mu$T lies in the horizontal plane under an angle of 25° to the trap axis. The $B$-field is measured with a sensor near the vacuum chamber and active feedback is applied in three dimensions via current modulation of six auxiliary coils.

The ion is cooled to approximately 0.5 mK, close to the Doppler limit, on the dipole allowed $^2S_{1/2} \rightarrow ^2P_{1/2}$ transition assisted by a repumper laser near 935 nm. Fluorescence from the decay of the $^2P_{1/2}$ state is collected by a lens of $N/A = 0.27$ and imaged onto the electron-multiplying charge-coupled device (EMCCD) camera$^{44}$. This enables state detection via the electron shelving technique. The ion can be prepared in either the $|^2S_{1/2}, m = -1/2\rangle$ or the $|^2S_{1/2}, m = +1/2\rangle$ state using circular polarized beam near 370 nm pointing along the direction of the quantization axis. The 467 nm laser for excitation on the E3 transition lies in the radial plane and its beam waist is $26(3) \times 38(3) \mu$m at the ion. The power is stabilized to $6.0(2)$ mW, at which a Rabi frequency of $\Omega_{E3}/2\pi = 10$ Hz is reached. The light is frequency-shifted and pulsed using acousto-optic modulators.

The resonant rf coil, consisting of 27 turns wound at a diameter of 4.5 cm, is placed 5.5 cm above the ion. The resonance frequency of the coil is $\omega_{res}/2\pi = 3.5147(7)$ MHz and it is driven by a signal derived from a direct digital synthesizer (DDS), referenced to a stable 100 MHz signal from a hydrogen maser. At $B = 221$ $\mu$T, the resonance frequency between the $m$-levels in the $^2F_{7/2}$ state is $\omega_{rf}/2\pi = 3.52$ MHz, close to the resonance frequency of the coil. The achieved multi-level Rabi frequency is $\Omega_{E3}/2\pi = 33$ kHz. The ambient magnetic field is monitored throughout the measurement campaign via data acquired in the E3 servo sequence. Drifts are observed at the level of $\Delta B = 100$ nT, corresponding to a detuning $\Delta\omega_{rf}/2\pi = 1.6$ kHz. The frequency supplied to the coils is actively adjusted to remain in resonance with the frequency given by the linear Zeeman splitting. For this purpose, the resonance frequency as a function of magnetic field was calibrated to be $\omega_{rf}(B)/2\pi = [1.581(1.6)B + 0.0162(3)]$ in MHz. For further details on the experimental set-up, see refs. 30, 41, 44, 45.

Relating the lab frame to the SCCEF

The elements of the $c_{\mu\nu}$ tensors are dependent of the local laboratory frame. In order to compare our results with those from other experiments, we transform the components of the local $c_{mn}$ tensor to the sun-centered, celestial-equatorial frame (SCCEF). We follow a similar derivation as given in ref. 6.

The relation between the components of $c_{mn}$ in the lab frame and $c_{MN}$ in the SSCEF is given by

$$c_{mn} = \Lambda_m^M \Lambda_n^N c_{MN},$$

where $\Lambda$ is the Lorentz transformation matrix, which consists of rotations and boosts in the lab frame relative to the SCCEF. Our lab frame has its origin at a colatitude of
\( \chi = 37.7^\circ \) and a longitude of \( \lambda = 10.5^\circ \) (PTB Braunschweig, Germany). In the local coordinate system, \( \hat{x} \) points towards the east, \( \hat{y} \) points towards the north and \( \hat{z} \) points upward. The rotation matrix that maps the SCCEF coordinate frame to that of the lab is given by

\[
R = \begin{pmatrix}
-\sin(\omega_\oplus T) & \cos(\omega_\oplus T) & 0 \\
-\cos(\chi)\cos(\omega_\oplus T) & -\cos(\chi)\sin(\omega_\oplus T) & \sin(\chi)
\end{pmatrix},
\]

where \( \omega_\oplus /2\pi = 1/23.934 \) h is the angular frequency of a sidereal day. Our quantization magnetic field \( B \) lies in the \( xy \) plane and points 20° south of east. Similar as in ref. 6, we calculate a virtual location on Earth’s surface, where \( B \) points vertically upward by only changing the origin of the coordinate system, not affecting transformation formulas between the lab frame and the SCCEF frame. This yields \( \chi_n = 102.1^\circ \) and \( \lambda_n = 84.4^\circ \). Substituting \( \chi = \chi_n \) and \( \lambda = \lambda_n \) values in equation (7) and setting \( T = 0 \) at the moment the \( z \) axis of the new location points towards the Sun on the day of the Vernal equinox, in this case 03:59:24 UTC, March 20, 2021 yields the proper transformation equations.

The boost of the experimental frame as seen from the SCCEF is given by

\[
\beta = \begin{pmatrix}
\beta \sin(\Omega_\odot T) \\
-\beta \cos(\eta)\cos(\Omega_\odot T) \\
-\beta \sin(\eta)\cos(\Omega_\odot T)
\end{pmatrix},
\]

where \( \Omega_\odot /2\pi = 1/(365.256 \times 24) \) h is the angular frequency of a sidereal year, \( \beta = 1 \times 10^{-4} \) is the magnitude of the boost from the orbital velocity and \( \eta = 23.4^\circ \) is the angle between the ecliptic plane and the Earth’s equatorial plane. Here the boost from the Earth’s rotation has been neglected as it is two orders of magnitude smaller (\( \beta_L = 1.5 \times 10^{-6} \)). Using the rotations and boosts, the Lorentz transformation that maps \( c_{\mu \nu} \) from the SCCEF to the lab frame is given by

\[
\Lambda = \begin{pmatrix}
1 & -\beta^1 & -\beta^2 & -\beta^3 \\
-(R \cdot \beta)^1 & R^{11} & R^{12} & R^{13} \\
-(R \cdot \beta)^2 & R^{21} & R^{22} & R^{23} \\
-(R \cdot \beta)^3 & R^{31} & R^{32} & R^{33}
\end{pmatrix},
\]

Using equation (9), the parameter \( C^{(2)}_0 \) can now be expressed in terms of components of \( c_{M,N} \) in the SCCEF using the Lorentz transformation. It is given by

\[
C^{(2)}_0 = A_0 + \sum_j [C_j \cos(\omega_j T) + S_j \sin(\omega_j T)],
\]

where \( A_0 \) is a constant offset, \( \omega_j \) contains all linear combinations of \( \omega_\oplus \) and \( \Omega_\odot \), and \( C_j \) and \( S_j \) are the respective amplitudes as given in Tab. S1.

With the high-pass filter applied to the data, we are sensitive only to signals that oscillate at frequencies larger than \( \nu_c = 5 \) \( \mu \)Hz. Therefore the Lorentz violating signal is given by

\[
C^{(2)}_0 = -3 \sin(2\chi)c_{XZ}\cos(\omega_\oplus T) - 3 \sin(2\chi)c_{YZ}\sin(\omega_\oplus T) - 3(c_{XX} - c_{YY})\sin^2(\chi)\cos(2\omega_\oplus T) - 3c_{XY}\sin^2(\chi)\sin(2\omega_\oplus T).
\]

10
\[
\begin{align*}
\omega_j & \quad C_j & \quad S_j \\
\omega & \quad -3 \sin(2\chi)c_{XZ} + 2c_{TY}\beta_L & \quad -3\sin(2\chi)c_{YZ} - 2c_{TX}\beta_L \\
2\omega & \quad \frac{-3}{2}(c_{XX} - c_{YY})\sin^2(\chi) & \quad -3c_{XY}\sin^2(\chi) \\
\Omega & \quad -\frac{1}{2}\beta(3\cos(2\chi) + 1)(c_{TY}\cos(\eta) - 2c_{TZ}\sin(\eta)) & \quad \frac{1}{2}\beta c_{TX}(3\cos(2\chi) + 1) \\
2\Omega & \quad 0 & \quad 0 \\
\Omega - \omega & \quad \frac{3}{2}\beta c_{TX}\sin(\eta)\sin(2\chi) & \quad -\frac{3}{2}\beta \sin(2\chi)[c_{TY}\sin(\eta) + c_{TZ}(1 + \cos(\eta))] \\
\Omega + \omega & \quad \frac{3}{2}\beta c_{TX}\sin(\eta)\sin(2\chi) & \quad -\frac{3}{2}\beta \sin(2\chi)[c_{TZ}(1 - \cos(\eta)) - c_{TY}\sin(\eta)] \\
2\Omega - \omega & \quad 0 & \quad 0 \\
2\Omega + \omega & \quad 0 & \quad 0 \\
\Omega - 2\omega & \quad -3\beta c_{TY}\cos^2(\eta/2)\sin^2(\chi) & \quad -3\beta c_{TX}\cos^2(\eta/2)\sin^2(\chi) \\
\Omega + 2\omega & \quad 3\beta c_{TY}\sin^2(\eta/2)\sin^2(\chi) & \quad -3\beta c_{TX}\sin^2(\eta/2)\sin^2(\chi) \\
2\Omega - 2\omega & \quad 0 & \quad 0 \\
2\Omega + 2\omega & \quad 0 & \quad 0 
\end{align*}
\]

**Table S1** The angular frequencies and the corresponding amplitudes contributing to \(C^{(2)}_0\) in the SCCEF as a function of the colatitude \(\chi\) and the angle between the ecliptic plane and the Earth’s equatorial plane \(\eta\).
Combining equation (11) with the sensitivity of $\kappa_{LV}$ to $C_0^{(2)}$ yields

$$\kappa_{LV} = 2\pi \times 5.1 \times 10^{15} \times [-3\sin(2\chi)c_{XZ}\cos(\omega_T)T - 3\sin(2\chi)c_{YZ}\sin(\omega_T)T - \frac{3}{2}(c_{XX} - c_{YY})\sin^2(\chi)\cos(2\omega_T)T - 3c_{XY}\sin^2(\chi)\sin(2\omega_T)T].$$

(12)

The stability of $\kappa_{LV}$ was measured to be $\sigma_\kappa = 372(9)$ mrad $\tau^{-1/2}$, from which we can extract the stability of $C_0^{(2)}$ to be $\sigma_{C_0^{(2)}} = 1.16 \times 10^{-17} \tau^{-1/2}$.

**The measurement sensitivity**

The quadratic term in the free Hamiltonian for state $|J, m\rangle$ interacting with a magnetic field $B = B_z \hat{z}$ is given by $H_{quad} = \kappa J_z^2$. It scales with $\kappa = \kappa_q + \kappa_{LV}$, where the first term is given by the quadrupole shift and the second term is given by a potential LV. The quadrupole shift can be calculated using

$$\Delta \nu_{quad} = \frac{1}{4} \frac{J(J + 1) - 3m^2}{J(2J - 1)} \frac{1}{\hbar} \Theta(2F_{7/2}) \frac{dE}{dz} [3\cos^2(\beta) - 1],$$

(13)

The quadrupole moment of the $^2F_{7/2}$ state is given by $\Theta(2F_{7/2}) = -0.0297(5)e a_0^3$, where $e$ is the electron charge and $a_0$ is the Bohr radius. The electric field gradient is given by $dE/dz = -m_{ion}\omega_z^2/q$ for a single trapped ion with mass $m_{ion}$ and charge $q$ at the equilibrium position of the trap. The angle between the quantization axis and the principle axis of the trap is in our case $\beta = 25^\circ$. The corresponding value for $\kappa_q$ is given by

$$\frac{\kappa_q}{2\pi} = -\frac{3}{4J(2J - 1)} \frac{1}{\hbar} \Theta(2F_{7/2}) \frac{dE}{dz} [3\cos^2(\beta) - 1].$$

(14)

At typical values of the axial secular frequency in our trap $\omega_{ax}/2\pi = (200 - 290)\text{kHz}$, the quadrupole shift is $\nu_{quad} = 60 - 125\text{mHz}$, corresponding to $\kappa_q = 0.075 - 0.150\text{rad/s}$. The fraction $P_f$ retrieved back into the $m = \pm 1/2$ state at the end of the rf sequence is dependent on the acquired phase $\kappa T_D$, where $T_D$ is the Ramsey dark time. At $\kappa T_D = 0.15$ rad, the slope $|dP_f/d\kappa|$ is maximum and the highest measurement sensitivity is reached.

**Fig. S1** The sensitivity of the measured quantity $P_f$ to variations in $\kappa$. a The calculated final population $P_f$ as a function of $\kappa$ at $T = 1.15\text{s}$, where the experimentally achieved contrast of 0.77(6) was taken into account. b The derivative $dP_f/d\kappa$ as a function of $\kappa$ at $T = 1.15\text{s}$. The measurement sensitivity at $\kappa_q = 0.13$ (green vertical line in a and b) is given by $dP_f/d\kappa|_{\kappa=0.13} = -4.4$. 

The sensitivity of the measured quantity $P_f$ to variations in $\kappa$. a The calculated final population $P_f$ as a function of $\kappa$ at $T = 1.15\text{s}$, where the experimentally achieved contrast of 0.77(6) was taken into account. b The derivative $dP_f/d\kappa$ as a function of $\kappa$ at $T = 1.15\text{s}$. The measurement sensitivity at $\kappa_q = 0.13$ (green vertical line in a and b) is given by $dP_f/d\kappa|_{\kappa=0.13} = -4.4$. 

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In the experiment, $\kappa = 0.13(3)$ for which an optimum Ramsey dark-time of $T_D = 1.15$ s is found. Using the average achieved contrast of $(P_{t,max} - P_{t,min})/(P_{t,max} + P_{t,min}) = 0.77$, $P_t$ and $dP_t/d\kappa$ are calculated as a function of $\kappa$, as shown in Fig. S1 a and b, respectively. At $\kappa_q = 0.13(3)$, the sensitivity of $P_t$ to variations of $\kappa$ is calculated to be $dP_t/d\kappa|_{\kappa=0.13(3)} = -4.4(4)$. The uncertainty on $\kappa_{LV}$ stemming from $\Delta(dP_t/d\kappa|_{\kappa=0.13(3)}) = 0.4$ is added to $\Delta\kappa_{LV}$ in quadrature.

**Data handling**

The E3 servo sequence is based on four measured populations at half the linewidth of the two opposite Zeeman transitions $|2S_{1/2}, m = \pm 1/2\rangle \rightarrow |2F_{7/2}, m = \pm 1/2\rangle$. The average value of the four measured data points are equal to $p_{E3}/2$, where $p_{E3}$ is the excitation probability of the E3 transition. Via the E3 servo sequence, repeated every 50 data points, $p_{E3}$ is monitored throughout the measurement campaign.

Slow drifts of $p_{E3}$ are observed on timescales of $\tau < 2.5$ days, corresponding to Fourier frequencies of $\omega/2\pi < 5 \mu$Hz, due to changes in, e.g., beam pointing and ambient noise. The quantity $P_t$ is detected via de-excitation from the $2F_{7/2}$ on the E3 transition and it is, therefore, highly correlated with $p_{E3}$. To quantify the correlation, data points are averaged over a time span of about one day and Pearson’s correlation factor is calculated to be 0.9. The number of measurements per averaged data point are not equal for $p_{E3}$ and $P_t$ and, therefore, the standard deviation of the two data sets are significantly different. For visualization purposes, the measured quantities are scaled by their respective standard deviation and plotted together with the 95% confidence interval, see Fig. S2. Note that Pearson’s correlation factor differs from 1, because the $p_{E3}$ and $P_t$ are not measured at exactly the same time, but rather in an alternating fashion.

The measured data points $P_t$ are corrected for slow drifts of $p_{E3}$. Residual slow variations that are not clearly connected to $p_{E3}$ are observed in the data at $\omega/2\pi = 1.65 \mu$Hz, related to a fluctuation on the timescale of a week. This Fourier component is removed from the data with a high-pass filter using a Hamming window with a cut-off frequency of $\nu_c = 5 \mu$Hz. Bounds on LV at frequencies $\omega_{\oplus}$ and $2\omega_{\oplus}$ are extracted from the filtered

![Fig. S2](image-url) Correlation between the E3 excitation probability $p_{E3}$ and the measured quantity $P_t$ averaged over a time span of about one day. The data points show $p_{E3}/\sigma_{E3}$ and $P_t/\sigma_t$ after subtracting the offset of 22.6 and 9.2, respectively. Pearson’s correlation is calculated to be 0.90. The ellipse indicates the 95% confidence interval.
data using a fit to equation (5).

To investigate the possible influence from the filter on the result, data points are simulated with Fourier components at \( \omega/2\pi = 1.5 \mu \text{Hz} \), \( \omega = \omega_\oplus \) and \( \omega = 2\omega_\oplus \), mimicking the observed slow drifts and a hypothetical LV signal, respectively. Exactly the same time stamps are used for the simulated data as for the experiment. The high-pass filter is applied to the simulated data with different values of \( \nu_c \), after which it is fitted to extract the amplitudes at \( \omega_\oplus \) and \( 2\omega_\oplus \). The retrieved amplitudes do not significantly differ from the simulated amplitudes for cut-off frequencies between \( 2 < \nu_c < 10 \mu \text{Hz} \). To validate this, the actual experimental data is also filtered with different values of \( \nu_c \) in the range of \( 2 < \nu_c < 10 \mu \text{Hz} \). The extracted amplitudes at \( \omega_\oplus \) and \( 2\omega_\oplus \) from the fit to equation (5) do not show a significant deviation for different values of \( \nu_c \) in this range.

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