Precision Measurement of Atomic Isotope Shifts Using a Two-Isotope Entangled State

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(Received 17 July 2019; published 11 November 2019)

Atomic isotope shifts (ISs) are the isotope-dependent energy differences between atomic electron energy levels. These shifts have an important role in atomic and nuclear physics, and have been recently suggested as unique probes of physics beyond the standard model under the condition that they are determined significantly more precisely than the current state of the art. In this Letter, we present a simple and robust method for measuring ISs by taking advantage of Hilbert subspaces that are insensitive to common-mode noise yet sensitive to the IS. Using this method we evaluate the IS of the $5S_{1/2} \leftrightarrow 4D_{5/2}$ transition between $^{86}Sr^{+}$ and $^{88}Sr^{+}$ with a 1.6×10^{-11} relative uncertainty to be 570 264 063.435(5)(8) (statistical)(systematic) Hz. Furthermore, we detect a relative difference of $3.46(23) \times 10^{-8}$ between the orbital g factors of the electrons in the $4D_{5/2}$ level of the two isotopes. Our method is relatively easy to implement and is indifferent to element or isotope, paving the way for future tabletop searches for new physics, posing interesting prospects for testing quantum many-body calculations, and for the study of nuclear structure.

DOI: 10.1103/PhysRevLett.123.203001

Despite its success, the standard model (SM) of physics has known limitations: it leaves cosmological phenomena such as dark matter [1,2], dark energy [2,3], and matter-antimatter asymmetry [2,4] unexplained, and it presents puzzles related to naturalness, such as the strong CP and hierarchy problems [2,5]. A central branch of research in physics attempts to address these problems by theoretically extending the SM and experimentally searching for signatures of such extensions. In the past several years, precise spectroscopic measurements of atomic and molecular systems have yielded strong constraints on new physics (NP) beyond the SM [6]; examples include searches for CPT violations [7], anomalous electron and nucleon electric dipole moments [8], local Lorentz invariance breaking [9,10], and time variation of natural constants [9]. Recently, a novel proposal has raised the possibility of detecting a new fundamental force beyond gravity and those of the SM using precision spectroscopy of isotope shifts (ISs) [11,12]. A new force of this sort can solve elementary problems in high-energy physics [13].

A force that couples neutrons to electrons will generate an isotope-dependent energy shift. Yet, this shift cannot be easily differentiated from SM contributions to the IS, which are difficult to calculate or measure independently. The proposal consists of an attempt to solve this problem by comparing several ISs. There are two dominant contributions to an IS: the mass shift (MS), arising from isotopes' different nuclear mass, and the field shift (FS), stemming from their different nuclear charge distribution. Each contribution can be approximated as a product of a nuclear (isotope-dependent) term and an electronic (transition-dependent) term. To the extent that this factorization holds, a linear relation can be drawn between ISs of different electronic transitions. This relation is known as King linearity [14,15]. The existence of a spin-independent fifth force that couples neutrons to electrons will add another contribution to the IS that, in general, will break King linearity. Hence, by comparing ISs and testing King linearity, one can place bounds on the parameters of new forces without necessarily knowing the exact values of the SM contribution to the IS.

However, reaching the spectroscopic precision necessary to attain state of the art bounds on such new forces is quite challenging, as current calculations place the required uncertainty at < 1 Hz [11]. Naïvely, reaching such a precision would obligate measuring optical atomic transitions with a relative uncertainty of 10^{-15} or below, a demanding feat accessible only to few leading optical clock labs [16]. Moreover, meaningfully testing the King plot linearity entails measuring—at the very least—two optical transitions in four different isotopes, all of which should be known within 1 Hz. Due to these difficulties, state of the art comparisons bound King linearity only down to the $\sim$100 Hz range [17], which is insufficient for the improvement of existing bounds.

In quantum metrological scenarios such as that of IS measurements, it is often possible to take advantage of techniques that filter out noise but leave the signal intact, thereby increasing sensitivity. A common and effective technique is that of decoherence free subspaces (DFSs), in which one engineers entangled states that evolve according to the signal operators, while dynamics take place entirely within a subspace that is invariant (i.e., degenerate) with respect to noise operators. Uses include measurement of the electric quadrupole moment of atoms [18], low uncertainty frequency comparison of two ions [19], imaging

spectroscopy with 100 μHz precision [20], measurement of the magnetic interaction between electrons in separate atoms at a distance of several micrometers [21], and tests of Lorentz invariance [10].

In this Letter, we present and implement a simple and powerful technique for measuring isotope shifts using a DFS. Our method forgoes measuring the optical transition and instead probes the isotope shift directly. The measurement dynamics takes place inside a DFS that is inherently immune to noise terms common to both isotopes, yet sensitive to the IS. Magnetic field and laser-phase noise mitigation allows for a significant prolongation of the measurement coherence time (in our case, a 100-fold), resulting in a corresponding decrease in statistical uncertainty. Immunity to other systematic frequency shifts, such as an electric quadrupole, a second-order Zeeman, and blackbody radiation (BBR) shifts, entails a low systematic uncertainty budget with comparatively little effort. The method is easy to implement and is not necessarily limited to ions. It essentially requires loading both isotopes into a trap and addressing and measuring each clock transition. We demonstrate our method using $^{88}Sr^{+}$ and $^{86}Sr^{+}$ ions and achieve an absolute (relative) uncertainty of 9 mHz ($\Delta f/f \sim 10^{-11}$) for this IS measurement. Similar precision with direct measurement of the optical transition frequency would have required a $\sim 10^{-17}$ relative uncertainty. We also measure a difference in the orbital angular momentum magnetic susceptibility, g_L, between the two isotopes, which we attribute to their small mass difference. Employing a DFS for isotope shift measurements was previously suggested by Roos [22].

In order to measure the IS, $\delta\nu^i_{nm} \equiv \nu^i_n - \nu^i_m$, of transition i between states $|g\rangle$ and $|e\rangle$, and of isotopes m, n, we trap a single ion of each isotope in a single chain in a linear Paul trap. Figure 2(c) shows the fluorescence image of such a two-ion crystal. We then prepare the maximally entangled Bell state,

$$|\psi_i\rangle = \frac{1}{\sqrt{2}}(|g_m e_n\rangle + e^{i\phi_0}|e_m g_n\rangle). \quad (1)$$

Here $|ge\rangle = |g\rangle \otimes |e\rangle$ where the left (right) hand side represents isotope m (n) and ϕ_0 is an arbitrary initial phase. This state can be prepared using sideband pulses (see the Supplemental Material [23]). The energy difference between the two states in this superposition, $E_{g_m} + E_{e_n} - (E_{e_m} + E_{g_n}) = (E_{e_n} - E_{g_n}) - (E_{e_m} - E_{g_m}) = h(\nu^i_n - \nu^i_m) \equiv h\delta\nu^i_{nm}$, is exactly the isotope shift times the Planck constant h. Therefore, during free evolution for time τ these states will acquire a relative phase, leading to the final state

$$|\psi_\tau\rangle = \frac{1}{\sqrt{2}}(|g_m e_n\rangle + e^{i\phi_0 - i2\pi\delta\nu^i_{nm}\tau}|e_m g_n\rangle). \quad (2)$$

This is a Ramsey sequence: by measuring the evolving phase between two superimposed states, we can measure the energy difference between the states, which is the IS.

The acquired phase is measured by applying two $\pi/2$ pulses, each at the carrier frequency of one of the isotopes, followed by a parity measurement [24,25], from which the isotope shift is deduced (see the Supplemental Material [23]). States of the form presented in Eq. (2) are degenerate with respect to global magnetic field noise, laser phase noise and any other shift common to both isotopes, forming a DFS. The Ramsey time τ is limited only by the coherence time of this superposition, which due to the absence of magnetic field and laser phase noise is determined by the lifetime of the excited states $|e_m\rangle, |e_n\rangle$.

A simpler variant of this method, yet with a lower signal-to-noise ratio, can be implemented without the need for entanglement. Here, we prepare each isotope individually in an equal superposition and arrive at the two-ion state,

$$|\psi'_0\rangle = \frac{1}{2}(|g_m g_n\rangle + |g_m e_n\rangle + |e_m g_n\rangle + |e_m e_n\rangle). \quad (3)$$

After a time $\tau_1 \gg \tau_d$, where τ_d is the dephasing time, the $|g_m g_n\rangle$ and $|e_m e_n\rangle$ part of the superposition will undergo complete dephasing, resulting in the density matrix $\rho_{\tau_1} = \frac{1}{4}(|g_m g_n\rangle\langle g_m g_n| + |e_m e_n\rangle\langle e_m e_n|) + \frac{1}{2}|\psi_{\tau_1}\rangle\langle\psi_{\tau_1}|$. Hence, with probability $\frac{1}{2}$ the required state is generated, and with probability $\frac{1}{2}$ the phase information is lost. The dephased part will average to a phase-insensitive null background in the parity measurement, corresponding to a loss of half the signal (see Fig. 1).

Measurements of the IS of the electron orbitals are, in general, affected by the weak dependence of the magnetic susceptibility of the orbital angular momentum on the mass of the nucleus. For a Hydrogen-like atom, the orbital g factor is corrected to be $g_L \approx 1 - m_e/m_N$ where m_e is the electron mass and m_N is the nucleus mass [26,27]. For a

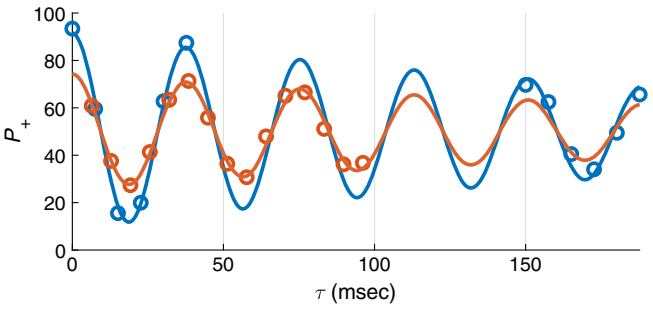

FIG. 1. The oscillation in time of the parity signal $P_+ \equiv P_{ee} + P_{gg}$ for the separable state and for the entangled state preparation sequence. The contrast of the entangled state sequence parity oscillation is almost twice that of the separable state sequence, due to the increased correlation of the entangled state. The oscillation frequency is determined by $\delta\nu^i_{88,86} - \delta f^i_{88,86}$, i.e., the difference between the isotope shift and the addressing fields frequency difference, which in this example is $\sim$26 Hz. Decay is due to the finite lifetime of the excited state. Data are shown in circles, while lines show fit as a guide to the eye.

many-electron atom, an additional correction due to correlations between electrons appears (see the Supplemental Material [23]). The differential magnetic susceptibility can be measured and eliminated by comparing measurements of different or opposite excited m levels and at different magnetic fields.

In our experiment, we measured the IS of the narrow (0.4 Hz) $|5S_{\frac{1}{2}}\rangle \leftrightarrow |4D_{\frac{5}{2}}\rangle$ electric-quadrupole transition between $^{88}Sr^{+}$ and $^{86}Sr^{+}$. Both clock levels were separated by a dc magnetic field of 3.5–5 G to their Zeeman sublevels which were split by several MHz. The transition was driven by a narrow (∼20 Hz) linewidth laser [28]. In order to resonantly address both isotopes, we split the laser into different AO frequency shifters that bridge the ∼570 MHz IS gap. The two beams were recombined and sent through another AO frequency shifter for common frequency control, and then passed to the ions through a single mode fiber to minimize differential optical phases along different paths.

After preparation of the Bell state $(1/\sqrt{2})(|D_{88}S_{86}\rangle + e^{i\phi_0}|D_{86}S_{88}\rangle)$ and a free evolution time τ, the phase difference between the closing $\pi/2$ pulses was scanned from 0 to 2π, giving a parity signal with some phase ϕ. This was repeated for two different times $\tau_i < \tau_f$. We extracted the IS frequency from the phase and time differences between the two sequences. As gradients of external fields, most prominently a gradient of the magnetic field, can generate an additional frequency difference between the two ions, we repeated the measurement for a reversed order of the two different isotopes in the crystal [Fig. 2(a)]. The average frequency of these two measurements was then added to our local oscillator frequency which was given by the difference of rf frequencies controlling the lasers for the isotopes. All rf sources were locked to a GPS-referenced high quality oscillator stabilized within 5×10^{-12} in one second. Averaging over many repetitions of this measurement yielded an isotope shift for the transition under some choice of the Zeeman levels m_S, m_D and the magnetic field B, which was measured independently. In order to determine the isotope shift at zero magnetic field, this process was repeated for Zeeman levels -m_S, -m_D, and at a different B. Combining all measurements we determined δg_L and $\delta\nu^{SD}_{88,86}$ using a maximum-likelihood analysis.

We performed three sets of measurements, under different combinations of external magnetic field B and Zeeman levels: $B \approx 5.17$ G, $m_S = 1/2$, $m_D = 3/2$; $B \approx 3.5$ G, $m_S = 1/2$, $m_D = 3/2$; and $B \approx 3.5$ G, $m_S = -1/2$ $m_D = -3/2$. For every set, we measured 2–4 traces, each consisting of 12–24 h of averaging [see Fig. 2(c)]. Traces of the same set were measured as far as three weeks apart demonstrating the system's stability. We calculated the Allan deviation [Fig. 2(b)] for each trace and found all traces to be shot noise limited; statistical uncertainty for a single trace varies from 9 to 15 mHz standard deviation.

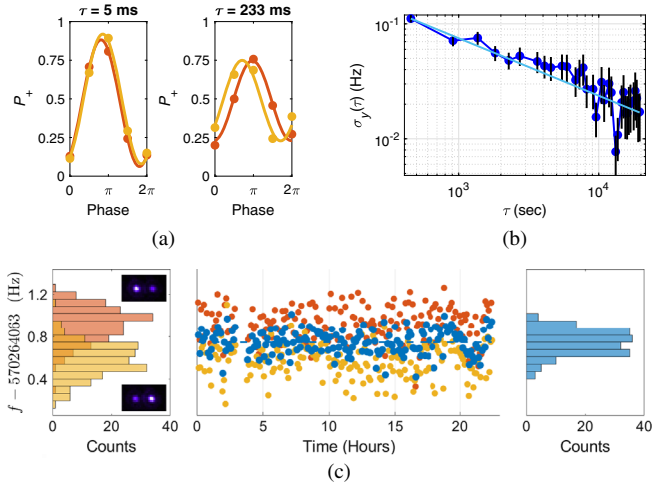

FIG. 2. A 24-hour IS measurement. (a) The superposition phase is determined for short (∼5 ms) and long (∼233 ms) interrogation times by varying the phase difference of the addressing fields and measuring the parity signal $P_+ = P_{SS} + P_{DD}$. From these the superposition oscillation frequency is extracted. The isotope crystal spatial configuration is alternated in order to average over external field gradients (color coded right and left). (b) Allan deviation analysis of data set shown in (c), consistent with shot-noise limit after 24 h of averaging. Mean values of every pair of alternate measurements is shown in blue. This measurement for $m_S = 1/2$, $m_D = 3/2$ with an external magnetic field of 5.17 G yields an oscillation frequency of 570264063.745 (9) Hz, which differs from the reported mean value of 570 264 063.435(9) Hz due to the isotope shift of the orbital magnetic susceptibility.

All traces are consistent with an electronic IS as well as an IS of the excited state magnetic susceptibility g_D, showing a linear dependence on the external magnetic field.

The main systematic shifts for optical ion clocks measuring at < 1 Hz uncertainty include the electric quadrupole shift, the BBR shift, the second-order Zeeman shift, and shifts due to the trap rf: an ac Stark shift and a second-order Doppler shift [16]. BBR, trap ac Stark shifts, and quadrupole shifts are common modes rejected up to the small isotopic changes to the electronic wave function (∼10^{-6}), rendering them negligible (<10^{-4} Hz). The differential second-order Zeeman shift is similarly negligible. Doppler shifts are only cancelled up to the mass difference between the ions, which provides a 1/40 reduction; however, as these shifts are relatively small to begin with (∼10 mHz), the mass reduction is sufficient to render them negligible as well.

Gradients of external fields can induce slow noise or constant systematic shifts. We mitigate their effect considerably by alternating the isotope positions and averaging over the measured shifts. However, the isotope position exchange is imperfect due to the mass dependence of the rf pseudopotential. Specifically, as the rf gradient is pronounced along the radial direction, isotopes that have exchanged axial positions can retain some radial offset due to uncompensated

radial dc fields and the mass difference. In this case, fields that have a non-negligible radial gradient over the offset distance will result in a systematic shift. We find that the residual rf field difference itself contributes a systematic shift [29] which we bound at 2 mHz, and the magnetic field radial gradient contributes a systematic shift, which we bound at 8 mHz, on par with our statistical uncertainty of 5 mHz. We measure the effect of each field independently (see the Supplemental Material [23]).

The final results of our measurement are shown in Fig. 3(a). Plotting the measured IS as a function of $B \times m_D$, we obtain a linear relation corresponding to a pure electronic isotope shift and a magnetic susceptibility isotope shift, generating the slope. From a maximum likelihood fit to a linear relation, we obtain an isotope shift at null magnetic field of $\delta\nu_{88,86}^{S,D} = 570\,264\,063.435(9)(5)(8)$ Hz (total)(statistical)(systematic), which corresponds to a relative uncertainty of 1.6×10^{-11}. Our uncertainty is a $\sim 10^{-17}$ fraction of the optical transition frequency. We also measure a difference in susceptibility of 0.0388(26) Hz/G, corresponding to a relative susceptibility isotope shift of $(\delta g_D^{5/2}/g_D^{5/2}) = \{[0.0388(26)][1.68 \times 10^6]\} = 2.31(15) \times 10^{-8}$, which translates to a relative susceptibility shift of the orbital angular momentum $(\delta g_L/g_L) = 3.46(23) \times 10^{-8}$ [30].

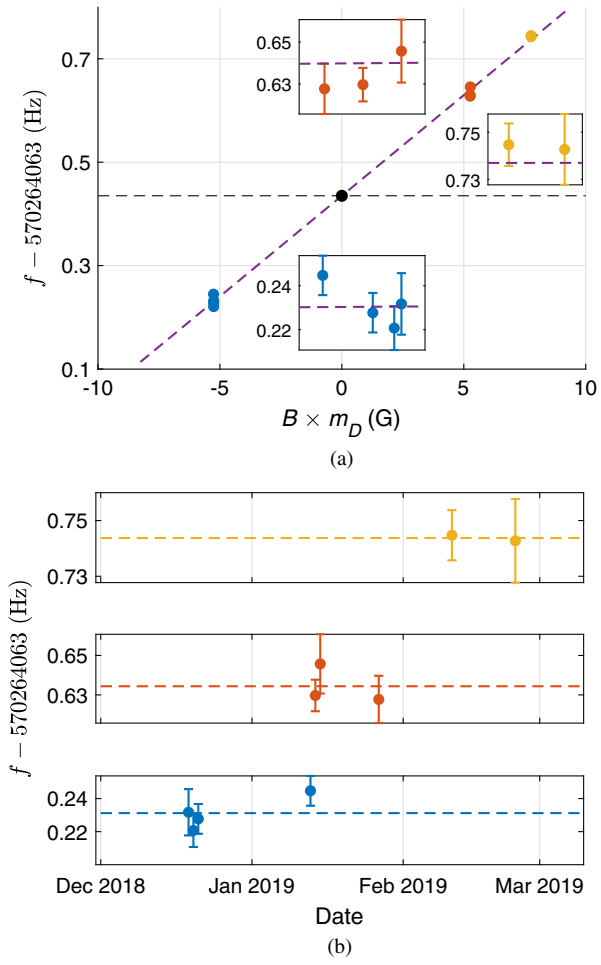

FIG. 3. Results summary: measuring $\delta\nu_{88,86}^{SD_{5/2}}$ and $\delta g_D^{88,86}$. (a) IS as a function of $B \times m_D$. Colors correspond to $B \approx 3.5$ G, $m_D = -3/2$; $B \approx 3.5$ G, $m_D = 3/2$; and $B \approx 5.17$ G, $m_D = 3/2$. Maximum likelihood linear fit shown in purple. The IS variation in $B \times m_D$ is due to g_D dependence on isotope, of which the slope of 0.0388(26) Hz/G is a direct measurement. The black dot denotes the IS at null magnetic field: 570 264 063.435(9) Hz. (b) IS over time. Measurements taken several weeks apart remain within the statistical uncertainty of a single measurement. Error bars represent statistical uncertainty of one standard deviation for all plots.

Discussion.—In order to test King linearity with sufficient accuracy to generate meaningful bounds on beyond-SM physics, at least six different IS measurements are necessary, including four different isotopes, all under 1 Hz uncertainty. Our method offers the high precision and simple adaptability needed in order to carry out these searches for new exotic physics. As illustrated here, uncertainties under 1 Hz are easily reachable using DFS for IS measurements. Even without the use of entanglement and careful suppression of stray fields, uncertainties under 100 mHz can be achieved within one hour of averaging. A King plot comparison with such low uncertainties would be a significant improvement on current state of the art [17,31].

The ability to interrogate optical isotope shifts with substantially improved precision presents a host of new opportunities for nuclear physics research. Due to their sensitivity to the nuclear charge radius, optical IS measurements are routinely used in order to probe nuclear structure [32–34]; in fact, optical IS measurements provide the most stringent bounds on nuclear charge radii for a large number of isotopes [35,36].

Precision IS measurements can serve as a test-bed for many-body atomic calculations. The specific mass shift (SMS) term requires evaluating the many-body electron correlation $\langle \sum_{ij} \vec{p}_i \cdot \vec{p}_j \rangle$, which is known to be difficult to calculate [37,38]. Using our method, the SMS can be resolved with an accuracy beyond what was available so far [39]. In addition to the SMS, we are able to measure the many-body correlation term $\langle \sum_{ij} \vec{r}_i \times \vec{p}_j \rangle$ through the specific orbital magnetic susceptibility, providing yet another independent test of the many-body wave function (for more details see the Supplemental Material [23]). This term is evaluated directly in the g factor IS measurement, as here there is no contribution to the nuclear finite charge radius complicating the analysis. Furthermore, our scheme can be used to investigate the isotope dependence of g_S, the magnetic susceptibility of the bound electron, by using the state $(1/\sqrt{2})(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle)$. Due to the long coherence time of this state, one can possibly reach the precision necessary for tests of QED and possibly beyond SM effects [40].

The two-isotope entangled superposition can be thought of as a synthetic rf IS atomic clock. Clocks use stable

periodic phenomenon in nature in order to keep track of time by counting periods. An IS clock is conceptually unique in the sense that, instead of using a local periodic phenomena, it uses nonlocal correlations between two oscillating subsystems as a periodic reference.

In summary, we present and demonstrate a novel method for measuring isotope shifts with trapped ions and use this method to measure the isotope shift between $^{88,86}Sr^{+}$ with high precision. The method makes use of the existence of a decoherence free subspace that is invariant to the most dominant noises, both fast and systematic. The method is simple and easy to use, as in essence it requires no more than carrier pulses and the measurement of two isotopes. Beyond measuring the IS of the optical transition, we determine the IS of the orbital magnetic susceptibility, which is sensitive to many-body electron correlations. Precision IS measurements open new possibilities in nuclear and atomic physics. Besides Paul traps, our method can be applied to metrology with optical tweezers [41]. The precision we demonstrate is far better than needed in order to potentially bound beyond SM physics (by repeating the experiment with several isotopes), and is the most precise optical IS measurement to date [42].

This work was supported by the Israel Science Foundation, the Israeli Ministry of Science Technology and Space, the Minerva Stiftung, and the European Research Council (consolidator Grant No. 616919-Ionology).

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