

# Probing the Atomic Higgs Force

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We propose an approach to probe new fundamental interactions using isotope shift spectroscopy in atomic clock transitions. As an example we focus on the Higgs boson couplings to the building blocks of matter: the electron and the up and down quarks. We show that the attractive Higgs force between nuclei and their bound electrons, that is poorly constrained, might induce effects that are larger than the current experimental sensitivities. We discuss how new interactions between the electron and the neutrons may lead to measurable non-linearities in a King plot comparison between isotope shifts of two different transitions. Given state-of-the-art accuracy in frequency comparison, isotope shifts have the potential of being measured with sub-Hz accuracy, thus potentially enabling the improvement of current limits on new fundamental interactions. Candidate atomic system for this measurement require two different clock transitions and four zero nuclear spin isotopes. We identify several systems that satisfy this requirement and also briefly discuss existing measurements. We consider the size of the effect related to the Higgs force and the requirements for it to produce an observable signal.

**Introduction** The Standard Model (SM) of elementary particles and fundamental interactions is one of the most successful scientific theories. Its last piece, the Higgs boson, was recently observed by the Large Hadron Collider (LHC) experiments [1, 2], a discovery of uttermost importance that led to the awarding of the 2013 Nobel Prize in Physics. Yet, the SM cannot be a complete description of nature. It does not possess a viable dark matter candidate, neutrino masses and mixings are unaccounted for, and it cannot explain the matter-antimatter asymmetry of our Universe. Furthermore, the masses of the charged fermions (quarks and leptons) exhibit a strong hierarchy, leading to the celebrated “flavor puzzle”.

Within the SM, every fermion mass  $m_f$  is induced by the product  $y_f \times v$ , where  $y_f$  denotes the fermion coupling to the Higgs boson, which corresponds to the strength of the Higgs force felt by the fermion  $f$ , and  $v = (\sqrt{2}G_F)^{-1/2} \approx 246 \text{ GeV}$  is the Higgs vacuum expectation value (VEV). Hence, the observed hierarchy of masses is a result of the *assumed* hierarchy in  $y_f$ , leading to a unique construct with

$$y_f^{\text{SM}} = \frac{m_f}{v}. \quad (1)$$

The Higgs discovery leads us to ask: is this new particle indeed the SM Higgs? It is possible that some of its

properties point to physics beyond the SM. An important new physics test is related to the Higgs boson couplings to the building blocks of matter: the electron and the up and down quarks, the proton’s and neutron’s “valence” quarks. We are currently rather ignorant regarding these, and within the SM these couplings (evaluated at the Higgs mass scale) are very small [3],

$$y_{e,u,d}^{\text{SM}}(m_h) \simeq 2.0 \times 10^{-6}, 5.4 \times 10^{-6}, 1.1 \times 10^{-5}. \quad (2)$$

In fact, it is possible that the strength of these Higgs-to-light-fermion interactions is far stronger than the above prediction [4], or that the light fermion masses are not due to the Higgs mechanism, resulting in much smaller couplings [5]. Furthermore, additional light degrees of freedom that are associated with the breaking of flavor symmetries and might even address the little hierarchy problem [6] may lead to a new scalar force with a larger coupling to the lighter charge fermions [7]. These cases lead to an alternative understanding of the flavor puzzle and to the establishment of new physics.

LHC Higgs data only directly probes the light quark couplings through the total Higgs width constraint  $\Gamma_h \leq 1.7 \text{ GeV}$  [8]. This translates into weak bounds of  $y_{u,d,s,c} \lesssim 0.3$  [9, 10], which is  $\mathcal{O}(10^4)$  larger than the SM values for  $u$  and  $d$  quarks. Global fits also indirectly constrain the light quark couplings, yet with additional assumptions on the production of the Higgs boson. Currently available LHC Higgs data together with electroweak (EW) precision tests yield a stronger bound of  $y_{u,d,s,c} \lesssim 1.6 \times 10^{-2}$  [9, 11, 12]. The electron coupling, on the other hand, is best probed through the direct search at the LHC of the  $h \rightarrow e^+e^-$  decay, giving  $y_e \lesssim 1.3 \times 10^{-3}$  [13, 14], see also [15]. The above

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bounds may be improved at next LHC runs. While direct bounds are not expected to significantly improve due to limitations in the detector resolution, indirect bounds from global fits could reach  $y_{u,d,s,c} \lesssim 5 \times 10^{-3}$  at the high luminosity stage [16]. The potential reach of the exclusive Higgs decays [12, 17, 18] to bound its couplings to constituent quarks is expected to be even weaker [16]. Meanwhile, the direct bound on the electron coupling could improve by an order of magnitude [15].

An alternative approach to the above experimental program at the energy frontier relies on low-energy precision measurements, for instance via atomic physics. Frequency measurements of narrow optical clock transitions in heavy atoms recently reached an unprecedented accuracy of  $\mathcal{O}(10^{-18})$  [19]. This remarkable level of precision paves the way for new tests of the existence of physics beyond the SM. Applications of atomic clock transitions have already been proposed in order to probe possible time-variation of fundamental constants [20, 21][20–22], and the existence of cosmological relics in the form of topological defects [23] or new ultralight particles [24] possibly associated with dark matter. We argue in this letter that sub-Hz precision measurements of isotope shifts in alkali or rare-earth atoms can potentially probe physics related to the origin of charged fermion masses. In this work we focus mostly on the physics of heavy force mediator. A discussion related to light mediators is to be reported elsewhere [25].

**Higgs force in atoms** Higgs boson exchange between a nucleus of mass number  $A$  and one of its bound electrons induces an attractive potential of Yukawa type, see for example [26],

$$V_{\text{Higgs}}(r) = -\frac{y_e y_A}{4\pi} \frac{e^{-r m_h}}{r}. \quad (3)$$

$m_h \approx 125 \text{ GeV}$  is the mass of the physical Higgs boson [27] and  $y_A = (A - Z)y_n + Z y_p$  is the effective nuclear coupling;  $Z$  is the atomic number and  $y_{n,p}$  are respectively the neutron and proton couplings. In terms of fundamental quark couplings (evaluated at the Higgs mass scale), they read [28–31]

$$\begin{aligned} y_n &\simeq 7.7 y_u + 9.4 y_d + 0.75 y_s + 2.6 \times 10^{-4} c_g, \\ y_p &\simeq 11 y_u + 6.5 y_d + 0.75 y_s + 2.6 \times 10^{-4} c_g, \end{aligned} \quad (4)$$

where  $c_g = 4.0 \times 10^2 y_c + 88 y_b + 1.5 y_t + \delta c_g$  is the effective coupling to gluons which includes the  $c, b, t$  contributions as well as a possible new physics contribution  $\delta c_g$ .

LHC data already indicate that the Higgs boson coupling to top and bottom quarks cannot deviate from the SM prediction by more than a factor few [32]. Given the direct bound above, the charm quark contributes at most  $\approx 0.03$  to  $y_{n,p}$  which is subdominant to the  $u, d, s$  contributions. Additional contributions to the Higgs-to-

gluon coupling are also constrained<sup>1</sup>,  $\delta c_g \lesssim \mathcal{O}(1)$  [32]. We therefore neglect  $c_g$  in the remainder. Within the SM, the  $u, d, s$  quark couplings are suppressed by the small fermion masses. Therefore, the heavy quarks dominate in Eq. (4), yielding small nucleon couplings,  $y_{n,p}^{\text{SM}} \sim 10^{-3}$ . However, requiring fundamental quark couplings to saturate the direct LHC constraints, nucleon couplings could reach values as large as  $y_{n,p} \sim 3$ ; while they are limited to  $y_{n,p} \lesssim 0.2$  by indirect bounds (see discussion above). Consequently, given the direct bounds on the quark and electron couplings, the strength of the Higgs force in atoms could be enhanced by a factor as large as  $10^6$  compared with the SM prediction.

The parity conserving part of the weak interaction constitutes another important effect, at least relative to the Higgs one mentioned above. The exchange of the  $Z^0$  boson contributes to the electron-nucleus potential at short-distance as

$$V_{\text{weak}}(r) = -\frac{8 G_F m_{Z^0}^2}{\sqrt{2}} \frac{g_e g_A}{4\pi} \frac{e^{-r m_{Z^0}}}{r}, \quad (5)$$

where  $g_e$  and  $g_A$  are the vector couplings of the  $Z^0$  boson to the electron and the nucleus, respectively; their tree level SM values are  $g_e^{\text{SM}} = -1/4 + s_W^2$  and  $g_A^{\text{SM}} = Q_W^{\text{SM}}/4$ .  $Q_W^{\text{SM}} = -(A - Z) + Z(1 - 4s_W^2)$  is the tree level SM value of the nuclear weak charge and  $s_W^2 \simeq 0.23$  is the sine of the weak mixing angle squared. While the electron  $Z^0$  coupling is known with  $\sim 10^{-3}$  accuracy from EW precision measurements at LEP [33], the corresponding couplings to first generation quarks are poorly constrained by data in a model independent way. In particular, the down-quark coupling to the  $Z^0$  may have a sizable deviation from its SM value [34], thus resulting in a significant uncertainty on the predicted weak nuclear charge. However, measurements of parity non-conservation (PNC) in atomic transitions can be used to accurately extract the value of  $Q_W$  at low energy. For example, in  $^{133}\text{Cs}$  atoms the agreement between the SM prediction and the experimental value is at the 0.5% level [35]. PNC measurements of similar precision in other heavy elements are also possible [36–39].

**Frequency shifts** We evaluate the Higgs contribution to atomic transition frequencies. Despite the possibly large nuclear Higgs coupling, the range of the Higgs interaction is extremely short, of  $\mathcal{O}(m_h^{-1}) \sim 10^{-3} \text{ fm}$ , and its strength remains much weaker than the dominant Coulomb interaction. The Higgs shift in electronic

<sup>1</sup> Sizable contributions to  $\delta c_g$  at the GeV scale could arise, while remaining invisible at the LHC, from a new physics sector which couples to QCD between the weak scale and the QCD scale. Such large contributions would however significantly modify the running of the QCD coupling and are therefore challenged by various precision measurements at low and high energies.

energy levels is then well-described in first-order (time-independent) perturbation theory. For the sake of simplicity, we derive our results using non-relativistic wave functions. In this limit,

$$\delta E_{nlm}^{\text{Higgs}} = \langle nlm | V_{\text{Higgs}} | nlm \rangle \simeq -\frac{y_e y_A}{4\pi m_h^2} |\psi(0)|^2 \frac{\delta_{l,0}}{n^3}, \quad (6)$$

where the ket  $|nlm\rangle$  is a solution of the Schrödinger equation for the unperturbed Coulomb potential, while  $n \geq 1$  and  $0 \leq l \leq n-1$  ( $-l \leq m \leq l$ ) are, respectively, the principal and angular momentum quantum numbers.  $|\psi(0)|^2/n^3$  is the wave-function-density at the origin ( $r=0$ ) for the electron of interest. In order to obtain  $\psi(0)$  we solve for the wave function including the presence of the inner shell electrons (see [40] for more details). We note that the transitions considered below are between partial waves of high angular momentum ( $D$ - or  $F$ -waves) with negligibly small overlap with the nucleus and an  $S$ -wave ( $l=0$ ) ground state. Hence, for this proposal, only  $S$ -wave energies are effectively shifted by the Higgs force. In the vicinity of the nucleus, the electron is typically in a relativistic regime and electron-electron interactions are important. A proper study of the relativistic and many-body effects in this region involves heavy numerical calculations of electronic structure, which are beyond the scope of this work. However, relativistic theory for electrons in atoms shows that non-relativistic wave functions yield a reasonably good estimate for  $S$ -waves around the nucleus [40, 41].

The frequency shift  $\Delta\nu = \Delta(\delta E)/2\pi\hbar$  resulting from the Higgs force for a  $n, l \rightarrow n', l'$  transition can be conveniently written as

$$\Delta\nu_{nl \rightarrow n'l'}^{\text{Higgs}} = 2.6 \times 10^2 \text{ Hz} \times y_e y_A \frac{|\psi(0)|^2}{4a_0^{-3}} I_{nn'}^{ll'}, \quad (7)$$

with  $a_0 \equiv (\alpha m_e)^{-1}$  is the Bohr radius,  $\alpha$  is the fine structure constant and  $I_{nn'}^{ll'} \equiv (\delta_{l,0}/n^3 - \delta_{l',0}/n'^3)$ .

Similarly, the frequency shift due to the weak interaction in Eq. (5), relative to the Higgs shift, is (assuming SM  $Z^0$  couplings)

$$\frac{\Delta\nu_{nl \rightarrow n'l'}^{\text{weak}}}{\Delta\nu_{nl \rightarrow n'l'}^{\text{Higgs}}} \simeq -4.9 \times 10^{-3} \frac{Q_W^{\text{SM}}}{y_e y_A}. \quad (8)$$

Equation (8) then indicates that the Higgs force is inevitably masked by the weak interaction, even with Higgs couplings saturating the conservative direct bound of  $y_e y_A \lesssim 0.004A$ . Therefore, the weak contribution must be known with relatively good accuracy, roughly given by Eq. (8), in order to probe Higgs couplings with precisely measured transition frequencies. Alternatively, assuming SM-like Higgs couplings, these measurements can also be recast into EW precision tests and provide complementary constraints on new physics through  $Q_W$ . Similarly to PNC measurements, the extraction of the weak nuclear charge would

also rely on accurate calculations of the atomic structure.

**Higgs shift in atomic clock transitions** The most accurate frequency measurements to date have been performed on narrow optical-clock transitions in laser-cooled atoms or ions, where state-of-the-art frequency comparisons are made with relative uncertainty in the  $10^{-18}$  range [19, 42]. Moreover, various spectroscopic investigations of optical-clock transitions in alkali-like systems are performed with sub-Hz accuracy [43–45]. We argue in the following that the Higgs-mediated contributions in these atoms are potentially much larger than the current collider bounds.

Consider for instance the optical electric-quadrupole  $nS_{1/2} \rightarrow n'D_{5/2}$  transition in  $^{88}\text{Sr}^+$  ( $Z=38, n=5, n'=4$ ) or in  $^{40}\text{Ca}^+$  ( $Z=20, n=4, n'=3$ ). We use Eq. (7) to estimate the expected frequency shift. In this case  $|\psi(0)|^2 \simeq 4.2(1+n_e)^2 Z/a_0^3$ , where the density of the valence electron at the nucleus approximately scales linearly with the nuclear charge  $Z$  (and not like  $Z^3$ ) due to the screening of core electrons [40]. We have included a factor of  $(1+n_e)^2$  relative to the result of [40] to account for the fact that in ions the typical radius of the valence electron is shorter by a factor  $\simeq 1+n_e$ , where  $n_e$  is the ion charge. Thus, the Higgs contributions could be as large as roughly 100 Hz and 50 Hz, respectively, with saturated bounds on the Higgs couplings. With reported accuracy of these transitions being below 1 Hz [43, 44], corresponding to a relative accuracy of  $\sim 10^{-15}$ , the experimental uncertainty on the evaluation of Higgs couplings would be of  $y_e y_{n,p} \lesssim 4 \times 10^{-5}$ , which is stronger than current collider (direct) bounds by a factor of  $\sim 100$ .

An even higher sensitivity to Higgs couplings can be obtained in  $\text{Yb}^+$  ( $Z=70$ ), where the Higgs shift is enhanced by the larger number of nucleons,  $A$ . A unique benefit of  $\text{Yb}^+$  is the presence of two narrow transitions in the optical range, namely the electric-quadrupole (E2)  $6S_{1/2} \rightarrow 5D_{3/2}$  and the electric-octupole (E3)  $S_{1/2}(4f^{14}6s) \rightarrow F_{7/2}(4f^{13}6s^2)$  transitions. Both transitions have also been recently measured with sub-Hz accuracy (0.36 Hz [46] and 0.25 Hz [21], respectively, see also [22]), yielding an uncertainty on extracting the Higgs coupling of  $y_e y_{n,p} \lesssim 4 \times 10^{-6}$ . Therefore, from an experimental point of view, the study of Higgs-mediated interactions in laser-cooled atoms seems very promising. On the theory side, the situation is much less promising. Indeed, the effect of many-body electron-electron interactions, along with different contributions that arise from the interaction of the valence electron with the nucleus, are not sufficiently known to be accounted for on the  $10^{-15}$  level.

**Isotope shifts** An alternative to comparison of absolute frequency measurements to theory would be to scrutinize frequency differences between several isotopes for the optical clock transitions. In principle these isotope shifts (IS) could also be measured with sub-Hz accuracy and their theory predictions are subject to less

uncertainties since the total charge  $Z$  remains constant. The Higgs contribution to the IS is roughly that of the individual transition frequency times the relative mass change between isotopes. For instance, in a frequency comparison between  $^{40}\text{Ca}^+$  and  $^{48}\text{Ca}^+$ , for the optical clock transition above, a change of  $\simeq 20\%$  in mass leads to a Higgs contribution to the IS of  $\sim 10\text{Hz}$  with saturated bounds ( $y_e y_n \simeq 0.004$ ). In a similar comparison between  $^{86}\text{Sr}^+$  and  $^{88}\text{Sr}^+$  ( $^{168}\text{Yb}^+$  and  $^{176}\text{Yb}^+$ ) a contribution of  $\sim 2\text{Hz}$  ( $\sim 10\text{Hz}$ ) is expected. IS in  $\text{Sr}^+$  were recently measured with a precision of  $\sim 4\text{kHz}$  [47]. Although experimental improvement down to the  $1\text{Hz}$  level for this very clock transition is realistic, theoretical calculations are still far from being able to predict the exact IS frequency in these atoms with such precision. In particular, the nuclear charge radius and many-body electron correlations typically result in large uncertainties. For instance, an *ab initio* frequency calculation by the authors of Ref. [47] disagrees with their measurement by more than  $20\%$ , with a discrepancy of  $\sim 100\text{MHz}$ .

**Breaking King's linearity** The IS between  $A$  and  $A'$  isotopes is usually thought of as arising from two different contributions: a mass shift (MS) and a field shift (FS) [48]. The MS is due to the nuclear mass change between the two isotopes. It receives contribution from a change in nuclear recoil (normal MS) and a change in electron-electron correlations (specific MS). Both effects are proportional to the relative mass change  $\mu_{AA'} \equiv 1/m_A - 1/m_{A'} = (A' - A)/(AA')\text{amu}^{-1}$ , where  $\text{amu} \approx 0.931\text{GeV}$  is the atomic mass unit. The FS, on the other hand, is due to the change in the charge distribution of the nucleus and it is approximately proportional to  $\delta\langle r^2 \rangle_{AA'}$ , the difference in the charge distribution variance between the two isotopes. Therefore, the IS for a given transition  $i$  is assumed to be of the form

$$\delta\nu_{AA'}^i \equiv \nu_{AA'}^A - \nu_{AA'}^{A'} = K_i \mu_{AA'} + F_i \delta\langle r^2 \rangle_{AA'}, \quad (9)$$

where  $K_i$  and  $F_i$  are, respectively, the MS and FS coefficients, that only depend on the transition, not on the isotopes. Both the specific MS and the FS pose a serious difficulty in calculating the IS from first principles as the change in nuclear charge radius and the proportionality factors in both cases are non-perturbative quantities.

A standard way to extract ratios and differences between the proportionality factors above, for two different transitions, and without knowledge of  $\delta\langle r^2 \rangle_{AA'}$ , is the King plot [49]. Defining modified IS as  $m\delta\nu_{AA'}^i \equiv \delta\nu_{AA'}^i/\mu_{AA'}$ , the change in charge radius between isotopes can be extracted from the IS in a single transition ( $i = 1$ ) as  $\delta\langle r^2 \rangle_{AA'}/\mu_{AA'} = (m\delta\nu_{AA'}^1 - K_1)/F_1$  and substituted in the IS expression for a second transition ( $i = 2$ ), which yields

$$m\delta\nu_{AA'}^2 = F_{21}m\delta\nu_{AA'}^1 + K_{21}, \quad (10)$$

with  $K_{21} \equiv (K_2 - F_{21}K_1)$  and  $F_{21} \equiv F_2/F_1$ . A linear relation between the (modified) IS associated with two

different transitions is therefore expected. If data are consistent with this linear relation, its slope  $F_{21}$  and offset  $K_{21}$  can then be extracted by plotting the IS of two transitions against each other for several isotope pairs.

With experimental accuracy below the Hz level, IS measurements will become sensitive to faint weak and Higgs contributions, in the presence of which Eq. (9) becomes

$$\delta\nu_{AA'}^i = K_i \mu_{AA'} + F_i \delta\langle r^2 \rangle_{AA'} + H_i (A - A'), \quad (11)$$

with  $H_i \equiv 2.7 \times 10^2 \text{Hz} \times (1 + n_e)^2 Z I_{nn'}^{U'} (y_e y_n - 4.9 \times 10^{-3} q_W^n)$  where  $q_W^n$  is the weak nuclear charge per neutron. In the SM,  $q_W^n = -1$  at tree level. The King relation in Eq. (10) is in turn modified as

$$m\delta\nu_{AA'}^2 = F_{21}m\delta\nu_{AA'}^1 + K_{21} - AA'H_{21}, \quad (12)$$

where we defined  $H_{21} \equiv (H_2 - F_{21}H_1)\text{amu}$ . Equation (12) shows that the Higgs and weak contributions explicitly break King's linearity law. A couple of comments are in order:

- Viewed from the atomic length scale, the finite nuclear size is characterized by a local interaction at the nucleus, like the Higgs and weak forces. Hence, to leading order,  $H_i \propto F_i \propto |\psi(0)|^2$ , which results in a vanishing  $H_{21}$  up to residual effects of the nuclear charge radius over the atomic radius, thus suppressing the sensitivity to Higgs couplings.<sup>2</sup> To our knowledge there is no precise calculation of  $H_i$  besides the above non-relativistic estimate, and we parameterize below the possible alignment between the  $F_i$  and  $H_i$  constants by a factor of  $S_{21} \equiv 1 - (F_2/F_1)(H_1/H_2)$ .
- There is a possibility for nature to accidentally conspire to cancel this non-linearity if  $m\delta\nu_{AA'}^i$  are linear functions of  $A'$ . In this case, the  $H_{21}$  term is a mere correction to the slope parameter  $F_{21}$  and sensitivity to any effect contributing to  $H_i$  is lost. While the precise isotopic dependence of  $m\delta\nu_{AA'}^i$  is straightforward to check directly from data, once available, we note that theory estimates strongly disfavour linear scaling of  $m\delta\nu_{AA'}^i$  with  $A'$ . This is expected because the charge radius of nuclei depends on their shell structure and therefore does not increase monotonically with the number of neutrons; see e.g. [50, 51]. We thus find these accidental cancellation to be unlikely.

It is possible therefore that, in the presence of new type of force mediator between the electron and nucleus, the King's law would be broken. Such an effect may be potentially observed in narrow optical clock

<sup>2</sup> We thank Krzysztof Pachucki and Maxim Pospelov for bringing this point to our attention.

transitions. Conversely, as long as IS data remains consistent with the King relation in Eq. (10),  $H_{21}$  can be bound largely independently of theory uncertainties. Furthermore, with sufficiently good knowledge of the atomic structure, in particular  $|\psi(0)|^2$ , and of the weak charge per neutron  $q_W^n$ , the  $y_e y_n$  combination of Higgs couplings can be constrained. State-of-the-art many-body simulations already predict the atomic structure of single-valence electron systems below the 1% level [35].

**The case of Yb ion** At least four isotopes are needed in order to probe the third term on the RHS of Eq. (12) through a deviation from linearity in a King plot. To this end, an appealing option is to use the two narrow optical clock transitions of  $\text{Yb}^+$ , namely the E2 and E3 transitions at 436 nm and 467 nm, respectively. In this case,

$$\frac{H_{21}^{\text{Yb}^+}}{\text{Hz amu}} \approx 0.9 \times (2.0 \times 10^2 y_e y_n - q_W^n) S_{21}. \quad (13)$$

The weak effect is expected at the Hz-level, while the Higgs force could appear slightly below 1 Hz under current constraints. The resulting sensitivity on the Higgs or other form of similar new physics can be estimated as follows. First of all, we assume that the weak contribution is subtracted from  $m\delta\nu$ 's with sufficient accuracy, and that a King plot constructed from the remainder IS shows a linear behavior. Thus, from Eq. (12),  $H_{21}$  is bounded to be smaller than the error on  $(m\delta\nu_{AA'}^2 - F_{21}m\delta\nu_{AA'}^1 - K_{21})/AA'$ , which we take to be dominated by the IS measurement uncertainty  $\Delta$ , yielding

$$y_e y_n \lesssim \frac{4 \times 10^{-4}}{|S_{21}|} \left( \frac{\Delta}{\text{Hz}} \right) \left( \frac{17Za_0^{-3}}{|\psi(0)|^2} \right) \left( \frac{8}{A' - A} \right). \quad (14)$$

As argued above, the reach of the method is suppressed in the limit  $S_{21} \rightarrow 0$ . While  $F_{21}$  could be extracted directly from the slope of the linear King plot, the ratio of  $F_{21}$  and  $H_2/H_1$  needs to be calculated. We note that despite the fact that a  $6S$  electron is active in both transitions, one may expect that at least  $1 - F_2/F_1 \sim \mathcal{O}(1)$ . The reason stems from the significantly different influence of core electrons between the E2 transition, where the  $4F$  shell is complete, and the E3 one, where it is missing one electron.

Combining Eq. (14) with Eq. (4), one obtains a sensitivity to the fundamental Higgs-to-light-quark couplings of

$$y_u + 1.2y_d + 0.10y_s \lesssim \frac{0.04}{|S_{21}|} \left( \frac{1.3 \times 10^{-3}}{y_e} \right) \left( \frac{\Delta}{\text{Hz}} \right), \quad (15)$$

neglecting the subdominant heavy quarks contribution.

Typical IS for clock transitions in  $\text{Yb}^+$  are in the GHz range, for example [52], while the experimental sensitivity is of  $\mathcal{O}(0.1\text{Hz})$ . Thus it is an important question to understand what is the expected size of the residual contributions from QED and the strong force, which were

neglected in Eq. (10). In particular whether these contributions are sufficiently suppressed and at most  $\mathcal{O}(10^{-9})$  relative to the leading terms. A parametric argument, in the non-relativistic limit, shows that non-linearities in a King plot induced by the nuclear effects are at least  $10^{-14}$  and  $10^{-10}$  smaller than the dominant IS contributions from FS and MS, respectively. We give below the general lines of the argument. First of all, observe that the IS is controlled by two small parameters: the difference of the electron reduced masses divided by their sum,  $\approx (m_e/2m_p)(1/A - 1/A') \simeq (A' - A)\varepsilon_\mu$  with  $\varepsilon_\mu \sim 10^{-8}$ , and the change in nuclear rms charge radius divided by the ion size,  $\langle \delta r^2 \rangle_{AA'} (\alpha m_e)^2 \sim (A' - A)\varepsilon_r$  with  $\varepsilon_r \sim 10^{-11}$ . The MS and FS in Eq. (10) are linear in  $\varepsilon_\mu$  and  $\varepsilon_r$ , respectively. Non-linear effects in the King plot could in principle arise at second order in these parameters, with size relative to the leading terms as large as  $(A' - A)\varepsilon_\mu \gg 10^{-9}$ . However, the ratio between the first and second order terms originating from the FS is independent of the transition up to corrections due to the overlap of the electron wave-function with the nucleus, resulting in an extra suppression of  $\mathcal{O}(\varepsilon_r)$ . Hence, non-linear effects from neglected FS corrections are at most  $\mathcal{O}[(A' - A)^2 \varepsilon_\mu^2] \sim 10^{-14}$ . There are other neglected effects from the specific MS. The leading contribution to the specific MS is  $\mathcal{O}(m_e \mu_{AA'})$ , while the sub-leading terms are  $\mathcal{O}[\alpha^2 m_e^2 (1/m_{A'}^2 - 1/m_A^2)]$  [53]. Therefore, non-linear effects from the MS are  $\mathcal{O}[\alpha^2 m_e (m_{A'} + m_A)/(m_A m_{A'})] \sim 10^{-10}$  level, which is small enough especially since the MS is typically sub-dominant to the FS for heavy nuclei [54]. We conclude that the breaking of King's linearity from residual QED and nuclear corrections is negligible. A more rigorous check of the negligibility of residual QED corrections should be performed using advanced atomic structure many-body, relativistic, calculations.

**Discussion** As a proof of concept, one can use the existing IS measurements in  $\text{Ca}^+$  ( $Z = 20$ ) for transitions involving the  $4S$  state [55]. With an error of  $\mathcal{O}(100)\text{kHz}$  and assuming  $S_{21} \sim \mathcal{O}(1)$ , this results in a rather weak bound of  $y_e y_n \lesssim 37$ . However, for light mediator we expect a much larger effect and this measurement might already lead to a meaningful bound [25]. As another example, radio-frequency E1 transitions were measured in Dy with very good accuracy [56] and with very good prospects for significant improvements [57]. A careful analysis of the electronic levels of Dy is however required in order to determine whether these systems are suitable for this purpose. Another possibility is to compare IS for clock transitions in an ion and its corresponding neutral atom, as done *e.g.* for Yb and  $\text{Yb}^+$  [52]. Since the nuclear parameters are the same for the ion and the neutral atom, the above analysis still holds. Therefore, additional Higgs-like forces can also be probed with non-linear King plots (as described by Eq. (12)) beyond the  $\text{Yb}^+$  case, using other systems like Ca, Sr and Hg, all of which have narrow clock transitions for the ion and the atom and at least four

stable isotopes [58]. We emphasise that this method can be rather effective in bounding new forces coupled to electrons and neutrons and whose range is comparable or longer than the typical nucleus size and can potentially lead to a stringent bound on the presence of light scalar and vector mediators [25].

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