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Probing New Long-Range Interactions by Isotope Shift Spectroscopy

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We explore a method to probe new long- and intermediate-range interactions using precision atomic isotope shift spectroscopy. We develop a formalism to interpret linear King plots as bounds on new physics with minimal theory inputs. We focus only on bounding the new physics contributions that can be calculated independently of the standard model nuclear effects. We apply our method to existing Ca data and project its sensitivity to conjectured new bosons with spin-independent couplings to the electron and the neutron using narrow transitions in other atoms and ions, specifically, Sr and Yb. Future measurements are expected to improve the relative precision by 5 orders of magnitude, and they can potentially lead to an unprecedented sensitivity for bosons within the 0.3 to 10 MeV mass range.

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Introduction.—The quest for new physics (NP) beyond the standard model (SM) of particle physics is pursued in multiple directions. Current efforts with colliders such as the LHC form the so-called energy frontier, probing directly the TeV energy scale. Other accelerators, such as meson factories, beam dump, and neutrino experiments, form the intensity frontier that broadly explores the MeV-GeV scale. Atomic physics tabletop experiments form a third frontier that broadly explores the MeV-GeV scale. Other accelerators, such as meson factories, beam dump, and neutrino experiments, form the intensity frontier that broadly explores the MeV-GeV scale.

atomic and molecular spectroscopy may probe spin-independent couplings of light boson fields to electrons and neutrons. The idea is to extract constraints from bounds on nonlinearities in a King plot comparison [16] of isotope shifts of two narrow transitions [17]. We develop a new formalism to interpret these measurements in the context of searching for new low-mass force carriers and propose several elements and transitions that can be used for such analyses. We recast existing measurements into bounds and provide an estimation for the sensitivity of future measurements; see Fig. 1. The validity of our method to bound NP does not rely on a knowledge of the SM contributions to King plot nonlinearities. Its constraining power, however, is limited by the size of the observed nonlinearities. In a case in which King linearity is established at the current state-of-the-art experimental precision—and barring cancellation between the SM and NP contributions—world-record sensitivity in a certain mass range will be achieved.

In this Letter we show that precision isotope shift (IS) spectroscopy may probe spin-independent couplings of light boson fields to electrons and neutrons. The idea is to extract constraints from bounds on nonlinearities in a King plot comparison [16] of isotope shifts of two narrow transitions [17]. We develop a new formalism to interpret these measurements in the context of searching for new low-mass force carriers and propose several elements and transitions that can be used for such analyses. We recast existing measurements into bounds and provide an estimation for the sensitivity of future measurements; see Fig. 1. The validity of our method to bound NP does not rely on a knowledge of the SM contributions to King plot nonlinearities. Its constraining power, however, is limited by the size of the observed nonlinearities. In a case in which King linearity is established at the current state-of-the-art experimental precision—and barring cancellation between the SM and NP contributions—world-record sensitivity in a certain mass range will be achieved.

FactORIZATION of IS. —Consider an atomic transition, i, between narrow atomic states. The difference in the transition frequency νi when comparing the isotopes
the coplanarity of the vectors. For each transition phenomenologically written as SN 1987A (light orange) [28], and star cooling in globular (397 vs 866 nm [19], the solid red line). IS projections (the specified on the labels). Constraint from existing IS data: Ca (for the experimental accuracies 10–17 10–14 10–11 0 10–5 10–2 10–1). The gray line at 17 MeV indicates the m med/med mediator mass. King linearity.

\[ N_L = \frac{1}{2} |(\vec{m}_{i1} \times \vec{m}_{i2}) \cdot \vec{m}_i| \quad (3) \]

In terms of the King plot, NL is the area of the triangle spanned by the three points shown in the Supplemental Material [37]. Equivalently, in the geometrical picture, it is half the volume of the parallelepiped defined by \( \vec{m}_{i1,2} \) and \( \vec{m}_i \). A given data set is considered linear if NL is smaller than its first-order propagated error

\[ \sigma_{NL} = \sqrt{\sum_k (\partial NL/\partial O_k)^2 \sigma_k^2} \]

where the sum runs over all measured observables \( O_k \) (modified frequency shifts and isotope masses) with standard deviations \( \sigma_k \).

New physics and violation of King linearity.—We now include a NP contribution by adding a third, also factorized, term to Eq. (1),

\[ \nu_i^{AA'} = K_i \mu_{AA'} + F_i \delta(r^2)_{AA'} + \alpha_{NP} X_i \gamma_{AA'} \quad (4) \]

where the two terms represent the mass shift and the field shift (FS), respectively [16,36]; \( \mu_{AA'} \equiv m_A^{-1} - m_{A'}^{-1} \), where \( m_A \) and \( m_{A'} \) are the masses of isotopes \( A \) and \( A' \), and \( \delta(r^2)_{AA'} \) is dominated by the difference in the charge radius of the two nuclei. Both \( \mu_{AA'} \) and \( \delta(r^2)_{AA'} \) are purely nuclear quantities that do not depend on the electronic transition \( i \), whereas \( K_i \) and \( F_i \) are isotope-independent, transition-dependent parameters. Given two electronic transitions, \( i = 1, 2 \), one obtains the following linear relation [16,36]:

\[ m_{i2}^{AA'} = K_{21} + F_{21} m_{i1}^{AA'} \quad (2) \]

with \( m_{i1}^{AA'} \equiv \nu_i^{AA'} / \mu_{AA'} \), \( m\delta(r^2)_{AA'} \equiv \delta(r^2)_{AA'} / \mu_{AA'} \), \( F_{21} \equiv F_2 / F_1 \), and \( K_{21} \equiv K_2 - F_2 K_1 \).

The formulas in our treatment of NP are simplified by introducing a geometrical description of the above leading-order (LO) factorization, as King linearity is equivalent to the coplanarity of the vectors. For each transition \( i \), we can form a vector \( \vec{m}_i \equiv (m_{i1}^{AA'}, m_{i2}^{AA'}, m_{i3}^{AA'}) \). The nuclear parameters of the field and mass shifts, \( \mu_{AA'} \) and \( \delta(r^2)_{AA'} \), can also be written as the vectors \( \vec{m}_i \equiv (1, 1, 1) \) and \( m\delta(r^2) \) in the same space, and hence Eq. (1) becomes

\[ \vec{m}_i = K_i \vec{m}_i + F_i m\delta(r^2) \quad (5) \]

where \( \vec{m}_i \) and \( m\delta(r^2) \) is illustrated in the Supplemental Material [37]. Like King linearity, coplanarity is a purely data-driven test of LO factorization since it is independent of theoretical input. A change in \( K_i \) and \( F_i \) will merely change the direction of \( \vec{m}_{i1} \) and \( \vec{m}_{i2} \) within the plane, but the qualititative statement of coplanarity remains. In this vector language we can provide a compact expression for a nonlinearity measure,
By solving the set of equations (4), one finds an expression for $\alpha_{NP}$ that is needed to yield a particular data set $\{\bar{m}v_1, \bar{m}v_2, \bar{m}\mu\}$,

$$\alpha_{NP} = \frac{(\bar{m}v_1 \times \bar{m}v_2) \cdot \bar{m}\mu}{(\bar{m}\mu \times \bar{h}) \cdot (X_1 \bar{m}v_2 - X_2 \bar{m}v_1)},$$

(6)

assuming that NP is the dominant contribution to nonlinearity. If linearity holds, then $\alpha_{NP} \lesssim \sigma_{\alpha_{NP}} = \sqrt{\frac{\Sigma_i (\partial \alpha_{NP}/\partial O_i)^2}{\sigma_i^2}}$. Hence, the sensitivity to $\alpha_{NP}$ is lost in the limit where the denominator in Eq. (6) vanishes because the NP contribution to nonlinearity is

$$NL_{NP} = \frac{\alpha_{NP}}{2} (\bar{m}\mu \times \bar{h}) \cdot (X_1 \bar{m}v_2 - X_2 \bar{m}v_1).$$

(7)

The presented method of limiting $\theta_{NP}$, Eq. (6), contains theory input only in $X_i$ and $h_{AA'}$, which describe how NP affects the IS. The SM contribution in the factorized limit is fully parametrized by the observables $\bar{v}_i$ and $\bar{m}$. The form of $h_{AA'}$ depends on the assumed couplings of new physics to nuclei. For example, if the new interaction couples to quarks, then we expect $h_{AA'} \propto AA$ [17,39]. The atomic transition-dependent factors $X_{1,2}$ can be determined by many-body simulation (see below).

Hence, in this method the background is estimated from data and only the NP contribution relies on theory input. This resembles the data-driven background estimation in collider searches for NP. As a consequence, precise predictions of the considered frequencies do not represent a selection criterion for suitable systems.

Thus far, most measurements of IS between spin-zero isotopes have been consistent with King linearity (see, however, the case of samarium in Ref. [40]). Nevertheless, some level of nonlinearity is expected to arise from SM higher-order contributions [41–44]. These contributions, which are related to nuclear physics and electronic-structure dynamics linked together, are presently not understood in a quantitative manner for many-electron systems. One possible source of nonlinearities is of the form of a field shift that depends on the electronic wave function with the nucleus. Hence, it might be possible to identify an observable that is less affected by the nucleus but is still sensitive to the presence of long-range NP interactions. In this regard, IS measurements involving highly excited, so-called Rydberg states might provide a smoking gun for the above types of NP.

For the proposed method to be effective, the element and the specific transitions should be chosen carefully. First, to make significant progress compared to the current precision, we consider narrow optical clock transitions. The most accurate frequency measurements to date, with a relative error of $10^{-18}$, have been performed on such transitions in laser-cooled atoms or ions [46–51]. Second, since the hyperfine interaction of electrons with the nucleus is a source for King nonlinearity [41], we consider only isotopes without nuclear spin.

**Contribution of new bosons to isotope shifts.**—Next, we discuss how theoretical IS predictions are modified in the presence of hypothetical new force carriers of spin $s = 0, 1, 2$ and mass $m_\phi$ which couple to electrons and neutrons with strengths $y_e$ and $y_n$, respectively. The effective spin-independent potential mediated by such bosons between the nucleus and its bound electrons is $V_\phi(r) = -\alpha_{NP}(A-Z)e^{-m_\phi r}/r$, where $\alpha_{NP} = (-1)^s y_e y_n/4\pi$. Note that NP could also couple to protons, though without affecting the linearity of the King plot; hence, we neglect such a coupling here. For concreteness, we consider $h_{AA'} = AA'$ amu for the NP contribution in Eq. (4).

The electronic NP coefficient $X_i$ can be determined via many-body calculations [52–58] at lowest order by calculating the overlap of the wave functions with the NP potential; see the Supplemental Material [37] for details.

We identify three regions of the NP interaction range, separated by the electron wave function size, $a_\phi/(1 + n_e)$, and the nuclear charge radius, $r_N \approx A^{1/3} \times (200 \text{ MeV})^{-1}$. Here, $a_\phi \approx (4 \text{ keV})^{-1}$ is the Bohr radius and $n_e$ is the ionization number. For $m_\phi \lesssim (1 + n_e)/a_\phi$, the “massless limit,” the interaction range is larger than the atomic size and $V_\phi \propto 1/r$, so $X_i$ becomes independent of $m_\phi$. For intermediate masses, $(1 + n_e)/a_\phi \lesssim m_\phi \lesssim 1/r_N$, the interaction range is within the size of the electron wave function, and the potential $V_\phi \propto e^{-m_\phi r}/r$ is mass dependent. Hence, detailed knowledge of the electronic wave functions is necessary to evaluate the effect of NP. In the heavy mass limit, $m_\phi \gtrsim 1/r_N$, the interaction range is shorter than the nuclear radius and $V_\phi \propto \delta(r)/(m_\phi^2 r^2)$. In this limit, the NP and nuclear charge-radius effects are approximately aligned since $X_i \propto F_i \propto |\Psi_b(0)|^2 - |\Psi_a(0)|^2$. This results
in a suppressed sensitivity for NP which scales as 

\((X_{21} - F_{21}) \to 0\); see the above and Ref. [17].

In the massless limit, \(X_i\) can be estimated without a
detailed computation of the atomic wave functions, as in
case the effective potential is Coulomb-like and thus its
effects are approximately accounted for by a shift of the
fine-structure constant \(\alpha\); see the Supplemental Material
[37] and Refs. [59,60]. We do not estimate the bounds on
theoretical value calculated in the absence of NP) has a
decreases approximately as

\(F_{\text{measured}}\) of indistinguishable from those of finite nuclear size. Bounds
are therefore suppressed by a factor of \(O(r_N/\alpha_0)\).

Current bounds and projections.—Here, we derive the
constraints on the product of electron and neutron coupling,
\(y_e y_n\), from existing IS data of \(\text{Ca}^+\) and project the
bounds for different transitions in alkaline systems in
the 10 eV–50 MeV mass range, assuming that better IS data
will be available in the future. Our results are summarized
in Fig. 1 and Table I.

We apply our method to the available IS data of \(\text{Ca}^+\)
(the solid line of Fig. 1). In the massless-boson limit, \(m_\phi \lesssim 10\) keV, the bound is essentially independent of \(m_\phi\).
At the high mass limit, we expect that \(F_{21} = X_{21}\). Since the
theoretical control of \(F_{21}\) is worse than the experimental
error, one can get an incorrect
bound at that limit. However, the ratio \(F_{21}^{\text{th}}/X_{21}\) (\(F_{21}^{\text{th}}\) is the
theoretical value calculated in the absence of NP) has a
much smaller error. Thus, in order to account for the
reduction in sensitivity as \(m_\phi\) increases, we rescale the \(y_e y_n\)
bound by \((1 - F_{21}^{\text{exp}}/X_{21})/(1 - F_{21}^{\text{th}}/X_{21})\), where \(F_{21}^{\text{exp}}\) is the
measured value of \(F_{21}\). We verified with the program
Grasp2K [58] that this factor does not change by more than a
few percent if the charge radius is changed by order 1.
(Since the latter is known to a few percent accuracy, this
is a rather conservative approach.) Indeed, we see that for
\(m_\phi > Z\alpha m_e\) the limits get weak, and the sensitivity
decreases approximately as \(m_\phi^{-3}\) for large masses. In the
Supplemental Material [37], we give two heuristic argu-
ments that obtain this asymptotic scaling of our loss of
sensitivity; the first is based on a perturbative approxima-
tion of \(X_i\), and the second is based on a nonrelativistic QED
effective theory [61–63].

For current bounds, we consider \(\text{Ca}^+\) (\(Z = 20\)). There
are five zero-nuclear-spin, stable, or long-lived isotopes
with \(A = 40, 42, 44, 46, 48\). References [19,64] reported
IS measurements for three isotope pairs \((A' = 42, 44, 48
relative to \(A = 40\)) in three dipole-allowed transitions in
\(\text{Ca}^+\) at wavelengths of 397.0 nm \((S_{1/2} - P_{1/2})\), 866.5 nm
\((D_{3/2} - P_{1/2})\) and 393 nm \((S_{1/2} - F_{3/2},\) not used here) with
an uncertainty of \(O(100)\) kHz.

Among alternative experiments that probe the \(m_\phi - y_e y_n\)
parameter space, we consider here only the ones most
sensitive to new light bosons coupled to electrons and
neutrons. The shaded regions in Fig. 1 summarize the
current reach of these experiments. We stress, however, that
some of them are derived involving further theoretical
assumptions, in contrast to our method, which relies on
few theoretical inputs. For new bosons lighter than a few
\(\times 100\) eV, fifth-force experiments [20,21] are potentially
sensitive. Since the interaction range covered by these
experiments is much larger than the atomic size, only forces
with nonzero atomic coupling can be probed. For illustra-
tion, we show in Fig. 1 the fifth-force bound applicable to
\(U(1)_{\mu - \tau}\) gauge bosons [18,65].

Furthermore, separately, \(y_n\) is constrained by various
neutron-scattering experiments [24–27], and \(y_e\) by the
anomalous magnetic moment of the electron \((g - 2)_e\)
[22,23] and by electron beam-dump experiments for \(m_\phi > 1\) MeV.

Both \(y_e\) and \(y_n\) are also severely constrained by globular
cluster energy loss for masses \(m_\phi < 350\) keV [28–33]
down to \(y_e y_n < 10^{-25}\) and \(y_e\) by sun cooling [66,67].
Couplings to nucleons in the \(\times 10^{-10} - \times 10^{-7}\) range for
\(m_\phi \lesssim 100\) MeV may be also excluded by energy loss in
the core of the supernova SN 1987A [28,68]. In order to
derive an upper bound on \(y_e y_n\), we combine for each mass
the best constraint on \(y_n\) from neutron experiments with \(y_e\)
either from \((g - 2)_e\) or from astrophysics.

As the precision of optical spectroscopy continues to
improve, higher accuracy IS measurements in different
systems can be achieved in the near future. Accordingly, we
estimate the sensitivity that would be achieved for several
transitions in alkali or alkali-earth ions or atoms, given
the improved accuracy. Here, we consider a comparison
between the two fine-structure split electric quadrupole
transitions in \(\text{Ca}^+\) and \(\text{Sr}^+\). A comparison between the
optical clock transitions in \(\text{Sr}^+\) and \(\text{Sr}\) and the quadrupole
and octupole transitions in \(\text{Yb}^+\) is also presented. In
principle, to enhance the sensitivity of our method, it is
desirable to compare transitions that involve levels that are
as different as possible. For this reason, comparing the two
fine-structure split electric quadrupole transitions in \(\text{Ca}^+\) or
\(\text{Sr}^+\) is not ideal, especially when compared to the sensi-
tivity of the two considered lines in \(\text{Yb}^+\) or comparing the
E2 line in \(\text{Sr}^+\) with the intercombination line in \(\text{Sr}^+\); see
Table I. We include these transitions in our projections.

<table>
<thead>
<tr>
<th>Transition 1 (nm)</th>
<th>Transition 2 (nm)</th>
<th>Accuracy</th>
<th>(y_e y_n) Bound ((m_\phi = 0))</th>
</tr>
</thead>
<tbody>
<tr>
<td>(\text{Ca}^+)</td>
<td>397.0</td>
<td>866.5</td>
<td>0.1 MHz</td>
</tr>
<tr>
<td>(\text{Ca}^+)</td>
<td>729.3</td>
<td>732.6</td>
<td>1 Hz</td>
</tr>
<tr>
<td>(\text{Sr}^+)</td>
<td>674.0</td>
<td>687.0</td>
<td>1 Hz</td>
</tr>
<tr>
<td>(\text{Sr}/\text{Sr}^+)</td>
<td>698.4</td>
<td>674.0</td>
<td>1 Hz</td>
</tr>
<tr>
<td>(\text{Yb}^+)</td>
<td>435.5</td>
<td>466.9</td>
<td>1 Hz</td>
</tr>
</tbody>
</table>
since their high-resolution IS measurement is experimentally simpler. All of the transitions above are expected to be measured with 1 Hz accuracy. Under the assumption that King linearity will hold in future measurements and following the Supplemental Material [37], the projected bounds are plotted in Fig. 1 (the dashed lines) as a function of \( m_\phi \) and summarized in Table I. The resonance structures at around the 10 keV scale arise from cancellations in the denominator of Eq. (6). These local losses of sensitivity at different masses per atomic system provide another motivation for IS measurements in complementary systems for a good coverage of the parameter space.

The various projections with 1 Hz accuracy significantly improve the bounds in the \( m_\phi \geq 10 \) keV region in parameter space. For lower \( m_\phi \) values, they are weaker than astrophysical bounds. However, astrophysical bounds are model dependent (for example, the chameleon effect [69]) and are subject to large uncertainties. Thus, an independent laboratory bound in this low-mass region is, nevertheless, worthwhile. For \( m_\phi \sim \) a few MeV, the projections of Ca\(^+\) (\( S \rightarrow D \) transitions) and Sr\(^+\) are comparable to the \( y_e y_n \) ones from neutron scattering [25] and \( (q - 2)_e \). Since neutron experiments are affected by uncertainties [27, 70–72], such as those involving the electron-neutron-scattering length, the nuclear input values, and the missing higher-order terms in the neutron-scattering cross section, the bounds in the high-mass range well above the neutron energies of \( E_n < 10 \) keV [25] should be understood as an indication of the order of magnitude. Consequently, theoretically, cleaner IS probes at the same order will already improve the bound robustness. Note that a Sr/Sr\(^+\) (Yb\(^+\)) IS comparison would become more effective than other existing methods in probing new bosons above \( \sim 10 \) keV, already with 100 Hz (1 kHz) accuracy (the bound related to Sr/Sr\(^+\) constructed from a comparison of transitions involving neutral and ion systems suffers from some numerical instabilities for masses above 20 MeV and is thus not shown). Finally, the range of \( y_e y_n \) needed to explain the Be anomaly [34,35] can be probed by future IS measurements of Yb\(^+\) at the 1 Hz level.

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See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.120.091801 for a visualization of the vector space.


