Letter of Interest for Snowmass 2021: Atomic/nuclear clocks and precision spectroscopy measurements for dark matter and dark sector searches

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Rapid development of atomic clocks and other precision spectroscopy techniques enables searches for bosonic dark matter and new force carriers. Improvements of many orders of magnitude in sensitivity are expected in the next decade. Strong interface of atomic and particle physics is essential for full realization of new opportunities presented by these advances.

Introduction. The past decade has been a transformative era for atomic, molecular and optical (AMO) physics. Revolutionary advances in the control of matter and light have enabled the precise interrogation and control of ultracold ions, atoms and molecules, and brought forth a wide variety of ultra-precise quantum sensors. This has resulted in a plethora of new AMO applications, including novel tests of the fundamental laws of physics. Precision low-energy measurements are a very powerful tool to probe new physics (NP) beyond the Standard Model (SM). Searches for NP with atoms and molecules have been recently reviewed in [1], including the topics of parity violation, searches for permanent electric dipole

moments, tests of the CPT theorem and Lorentz symmetry [2], searches for spatiotemporal variations of fundamental constants, tests of quantum electrodynamics, tests of general relativity and the equivalence principle, searches for dark matter, dark energy and extra forces, and tests of the spin-statistics theorem.

The unprecedented progress in accuracy is bound to have profound implications for experimental tests of fundamental forces, particularly in combination with new ideas to probe the physics of dark matter, new light force carriers, and scalar particles, as discussed in this letter. Here we focus on two promising examples: atomic clocks and precision spectroscopy. Both are sensitive to new light physics. A number of beyond the SM theories proposed to solve various problems of the SM, such as the gauge hierarchy, the strong CP problem or the flavor puz-

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zle, feature light new particles with masses well below the GeV scale and small couplings to the matter fields [3–12].

Clocks. Optical atomic clocks have improved by more than three orders of magnitude in precision in less than 15 years [13], reaching a fractional frequency precision of 10^{-18} . If the fundamental constants, such as the finestructure constant α or proton-to-electron mass ratio, are space-time dependent, so are atomic and molecular spectra and the clock frequencies. Since frequencies of different clocks depend differently on fundamental constants [1], their variation with the fundamental constants would change the clock tick rate and make it dependent on the location, time, or type of the clock. Earlier atomic clock searches focused on the "slow-drift" model of fundamental constant variation (on time scales from days to the lifetime of the Universe) and testing the coupling of fundamental constants to a changing gravitational potential [1, 13]. Recently, it was shown that ultralight scalar bosonic dark matter (DM) can source oscillatory and transient variation of fundamental constants ([14– 19], also see review [20]). In our Galaxy, such DM exhibits coherence behaving like a wave with an amplitude $\sim \sqrt{\rho_{\rm DM}}/m_{\rm DM}$, where $\rho_{\rm DM} = 0.3 \ {\rm GeV/cm}^3$ is the local DM density [14]. The coupling of such DM to the SM leads to oscillations of fundamental constants and, therefore, clock transition frequencies. Such oscillation signal would be detectable with atomic clocks for a large range of DM masses and interaction strengths. Recently, new bounds on the coupling of ultralight dark matter to SM particles and fields were set by conducting frequency comparisons between a state-of-the-art strontium optical lattice clock, a cryogenic crystalline silicon cavity, and a hydrogen maser [19].

Transient changes in fundamental constants that are potentially detectable with networks of clocks may be induced by the DM objects with large spatial extent, such as stable topological defects [15, 16, 21, 22]. A recent large-scale comparison between European optical clocks has improved bounds on transients of α [21, 22]. In summary, atomic clocks can map small fractional variations of α (of any cause or type, e.g., temporal, spatial, slow-drift, oscillatory, gravity-potential dependent, transient or other) onto fractional frequency deviations that are measured with outstanding precision.

It was also proposed that high-energy astrophysical events could produce intense bursts of exotic low-mass fields (ELFs) potentially detectable with networks of atomic clocks and other quantum sensors expanding the toolbox of multi-messenger astronomy [23]. Such precision quantum sensor networks can function as ELF telescopes to detect ELF bursts signals of sufficient intensity.

Several directions are being pursued to drastically improve the reach of the clock and other precision spectroscopy experiments for DM detection: (i) significant improvement of the current clocks [2], that is expected to rapidly evolve in the next decade; (ii) development of clock networks at the new level of precision [21, 22]; (iii) development of new atomic clocks based on highly charged ions (HCI) that have much higher sensitivities to the variation of α [24–26]; (iv) development of a nuclear clock that is based on a nuclear rather than atomic transition [27, 28]; (v) dedicated precision spectroscopy experiments sensitive to higher DM masses than clocks (WReSL experiment [29]); (vi) development and implementation of new clock-comparison schemes specifically designed to improves reach of oscillatory and transient dark matter searches [30]; and (vii) development of molecular clocks [31, 32]. The experimental effort is strongly complimented by the development of highprecision atomic theory [33] and particle physics model building [8–12].

Atomic clocks also provided the most stringent tests of Lorentz symmetry in the electron-photon sector [2]. Several orders of magnitude improvements of these tests are expected with novel proposed schemes (see the LoI on CPT and Lorentz symmetry tests).

HCI are attractive candidates for the development of novel atomic clocks with very high sensitivity to both dark matter and variation of α . The HCI optical clock proposals, fundamental physics applications, and experimental progress towards HCI high-precision spectroscopy were recently reviewed in [24]. Recent development of HCI cooling, trapping, and quantum logic techniques are enabling rapid progress in the development of HCI clocks [25, 26].

The transition frequencies of nuclear energy levels are generally outside of the laser-accessible range by many orders of magnitude. A single exception is a long-lived nuclear transition that occurs between an excited state (isomer) of the ²²⁹Th isotope and the corresponding nuclear ground state, with wavelength near 150 nm, within reach of modern lasers. The transition energy have been recently measured [27]. Designing a clock based on this nuclear transition [28] is particularly attractive due to the suppression of the field-induced frequency shifts as the nucleus interacts only via the relatively small nuclear moments and is highly isolated from the environment by the electron cloud. The nuclear clock sensitivity to the variation of α is expected to exceed the sensitivity of present clocks by ~ 4 orders of magnitude [34]. In addition, nuclear clocks will be sensitive to a DM coupling to the hadronic sector of the SM. The expected extreme sensitivity of the nuclear clock to the variation of the fundamental constants and related NP is based on the fact that the energy scales of the internal nuclear interactions are several orders of magnitude higher than the actual nuclear transition energy, which can be measured with a precision that is unprecedented in nuclear physics. At the projected 10^{-19} fractional frequency precision level and strongly enhanced sensitivity, the nuclear clock can improve the ability to probe scalar dark matter by 6-7 orders of magnitude for a wide range of DM mass ranges in comparison with present limits [17–19].

The WReSL experiment [29] searches for fast apparent oscillations of the fundamental constants with frequencies of up to 100 MHz. The idea is to use highresolution laser spectroscopy to detect fast oscillations of an atomic-transition frequency with a laser locked to an optical cavity with a different dependence on the oscillating constants. Indeed, a broad class of DM scalar fields manifest themselves as a variation of constants and in this context, it is legitimate to consider variations of dimensionful constants, which is not possible for the case of a slow drift [35]. Even tighter bounds have been achieved by comparing optical and microwave clocks to cavities [36, 37].

Isotope shifts. A straightforward method to probe NP is a direct comparison between the SM and NP theoretical predictions and the experimental results, either by absolute frequencies or by isotope shifts (IS). The latter are the difference of the same transition frequency in different isotopes and have the advantage of reduced uncertainties. However, such a direct comparison is possible only in the simplest systems, such as H, He atoms and positronium, where the SM contribution is known with accuracy better than the current experimental sensitivity see *e.g.* [38–43]. For example, the hydrogen-deuterium isotope shift is a competitive bound on a new electronneutron spin-independent interaction with a mediator mass at the MeV scale [40].

In more complicated systems, such as Ca, Sr, and Yb, the theoretical predictions are much less accurate than the experimental uncertainties. However, by using (approximate) symmetries or factorization properties of the system, we can combine experimental measurements to construct observables with much smaller theoretical uncertainties, resulting in an enhanced NP sensitivity. One such example is the King plot analysis of precision IS spectroscopy. By considering the SM leading contribution to the IS (mass and field shifts), it was shown that there is a linear relation between two electronic transitions (with respect to different IS measurements) [44]. New spin-independent interactions between the electron and the neutron will break this relation, and thus can be probed by looking for a deviation of the King plot from linearity [45, 46]. These spin-dependent interactions can be a result of tree level exchanges of new scalar or vector

bosons with a mass below $\mathcal{O}(50 \text{ MeV})$, which appear in different extensions of the SM. See Ref. [47] for interpretations in a context of several beyond the SM models. Particularly interesting, in this regard, is a class of singlet scalar portal models, where the scalar mixes with the Higgs [48]. This class of models is motivated by the relaxion framework [8] that effectively can be described as a finely tuned Higgs-portal model [10, 12, 49], and that may be potentially probed by the methods described in this LoI [50].

Higher-order SM contributions break the linearity of the King plot as well [46] and must either be calculated with high accuracy [51], or eliminated together with uncertainties on the isotope mass differences using more transitions and a generalized analysis [50, 52]. The most stringent bound on such fifth forces has been established based on Ca⁺ isotope shift spectroscopy with up to 20 Hz resolution [53], while Yb⁺ measurements have revealed a 3σ nonlinearity, possibly caused by higher-order contributions within the SM [54]. A possible improvement of the bounds can be achieved by choosing one transition in the King-plot comparison from a trapped-ion clock and the other from neutral atoms optical lattice clocks of the same species; e.g. Sr⁺ and neutral Sr clocks.

Future HCI isotope shift spectroscopy of e.g. Ca^{14+} and Ca^{15+} isotopes is a promising avenue since the fewelectron systems can be calculated with high accuracy and offer several narrow transitions for removing SM nonlinearities.

Complementarily, isotope shifts of Rydberg transitions allow to probe long-range interactions based on only two isotopes and the measured polarizability as an input [55].

Conclusions. Rapid developments of atomic clocks and other precision spectroscopy techniques are making possible phenomenologically interesting searches for bosonic dark matter and new force carriers. Furthermore, it is realistic to expect in the next decade many orders of magnitude improvements in the sensitivity of these experiments. To take full advantage of new opportunities presented by these advances will require experimental and theoretical engagements from both atomic and particle physics communities.

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