Search for Ultralight Dark Matter from Long-Term Frequency Comparisons of Optical and Microwave Atomic Clocks

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We search for ultralight scalar dark matter candidates that induce oscillations of the fine structure constant, the electron and quark masses, and the quantum chromodynamics energy scale with frequency comparison data between a ^{171}Yb optical lattice clock and a ^{133}Cs fountain microwave clock that span 298 days with an uptime of 15.4%. New limits on the couplings of the scalar dark matter to electrons and gluons in the mass range from 10^{-22} to 10^{-20} eV$/c^2$ are set, assuming that each of these couplings is the dominant source of the modulation in the frequency ratio. The absolute frequency of the ^{171}Yb clock transition is also determined as 518 295 836 590 863.69(28) Hz, which is one of the important contributions toward a redefinition of the second in the International System of Units.

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While the existence of dark matter is indicated by various astrophysical observations [1], its constituents have not been conclusively detected in laboratory experiments. In such experiments, dark matter candidates with particle physics motivation are studied particularly well. Weakly interacting massive particles in the mass range from 1 to 10^3 GeV$/c^2$ (c, speed of light) have been searched for in various experiments [2–7]. Quantum chromodynamics (QCD) axions and axionlike pseudoscalar particles and fields with masses of $\lesssim 10$ eV$/c^2$ have also attracted considerable attention. Limits have been set on the interactions between the axions and the standard model particles such as electrons, photons, gluons, nucleons, and antiprotons [8–13]. The range of the search broadens as different kinds of technologies are applied to dark matter searches.

Other possible candidates for dark matter are dilatonlike ultralight scalar fields with their masses m_φ far below 1 eV$/c^2$, down to 10^{-22} eV$/c^2$ required by de Broglie wavelength of the scalar field being smaller than the sizes of dwarf galaxies [14]. Such an ultralight bosonic field as dark matter has large occupation numbers per mode and behaves as a classical wave with a frequency proportional to m_φ [15]. There has been an astrophysical interest in the ultralight dark matter in the mass range from 10^{-22} to 10^{-21} eV$/c^2$, as its wave property might solve long-standing problems of cosmic small-scale structures [14,16,17]. If the scalar field couples to the standard model fermions and gauge bosons [18], it induces coherent oscillations of fundamental constants such as the fine structure constant α and the electron mass m_e [19]. The scalar field can also form macroscopic clumps called topological defects [20], leading to transient variations of fundamental constants by the passages of such clumps [21–25].

The variation of fundamental constants [26–36] and thus the existence of the ultralight scalar fields can be detected with frequency ratios of atomic clocks based on different atomic species and transitions. The periodic oscillations of the frequency ratios have so far been searched for by clock comparisons involving optical and microwave clocks with different sensitivities to fundamental constants. Previous comparisons between two optical clocks took advantage of the state-of-the-art short-term frequency stabilities [37] and accuracies [38–40] to search for α oscillations in the frequency range of $\lesssim 0.1$ Hz and set stringent bounds on coupling constants of the scalar fields to photons in the mass range of $m_\varphi \lesssim 10^{-16}$ eV$/c^2$ [41,42]. Frequency ratios of two microwave clocks are sensitive to the couplings to quarks and gluons as well as photons. Although microwave clocks have relatively low short-term stabilities, they are operated for long periods with high uptimes and contribute to searches in low frequency (i.e., mass) regions. A comparison between Rb and Cs fountain microwave clocks for six years yielded stringent limits on the couplings to quarks and gluons in the mass range of $m_\varphi \lesssim 10^{-21}$ eV$/c^2$ [43].

The unique feature of comparisons between optical and microwave clocks is the capability of the observation of the oscillation of m_e induced by the scalar field couplings to electrons [19]. The longest search for oscillations in the optical to microwave ratio was performed in a comparison between a Si optical cavity and a hydrogen maser (H maser) over 33 days [41]. Assuming that the scalar fields predominantly couple to electrons, this report put constraints on the coupling between the scalar field and m_e in the mass region from $m_\varphi = 10^{-21}$ to 10^{-18} eV$/c^2$. To extend the search to the lowest mass limit of 10^{-22} eV$/c^2$, longer measurement periods are required. It is also desirable to compare an optical lattice clock or a single ion optical clock with a fountain microwave clock to improve the dark matter detection sensitivity that is limited by the frequency instability of the Si/H measurement induced by flicker and random-walk frequency noises at long averaging times [44].

In this Letter, we report on a search for the oscillating scalar dark matter fields with frequency comparison data between a ^{171}Yb optical lattice clock and a ^{133}Cs fountain clock that span 298 days with an uptime of 15.4%. The main technical advantage in our search is the robustness of the Yb optical lattice clock, which can be operated with high uptimes for several months [45]. We establish improved constraints on the couplings of the scalar field to electrons and gluons in the mass range from $m_\varphi = 10^{-22}$ to 10^{-20} eV$/c^2$, assuming that each of these couplings is the dominant source of the oscillation. In addition to the dark matter search, we also provide the absolute frequency of the Yb clock transition, which is one of the important contributions toward a redefinition of the second in the International System of Units [46,47].

We consider linear couplings between the scalar field and the standard model particles described by the interaction Lagrangian [18,19,43]

$$\mathcal{L}_{\mathrm{Int}} = \varphi\left[\frac{d_\alpha}{4\mu_0}F_{\mu\nu}F^{\mu\nu} - \frac{d_g\beta_3}{2g_3}F^A_{\mu\nu}F^{A\mu\nu} - c^2\sum_{i=e,u,d}(d_{m_i}+\gamma_{m_i}d_g)m_i\overline{\psi_i}\psi_i\right], \tag{1}$$

where φ denotes a dimensionless scalar field relative to the Planck scale [18,43], $F_{\mu\nu}$ the electromagnetic tensor, μ_0 the magnetic permeability, $F^A_{\mu\nu}$ the gluon field strength tensor, g_3 the QCD coupling constant, β_3 the β function for the running of g_3, γ_{m_i} the anomalous dimension describing the running of the mass m_i of the QCD-coupled fermion, and ψ_i the fermion spinor. Five dimensionless coefficients d_α, d_{m_e}, d_{m_u}, d_{m_d}, and d_g are coupling coefficients of the scalar field to photons, electrons, up and down quarks, and gluons, respectively. It has been shown in Ref. [18] that these couplings result in the following linear dependence of the fundamental constants with respect to the scalar field:

$$\alpha(\varphi) = \alpha(1+d_\alpha\varphi), \qquad m_e(\varphi) = m_e(1+d_{m_e}\varphi),$$
$$m_q(\varphi) = m_q(1+d_{m_q}\varphi), \qquad \Lambda_{\mathrm{QCD}}(\varphi) = \Lambda_{\mathrm{QCD}}(1+d_g\varphi), \tag{2}$$

where Λ_{QCD} denotes the QCD energy scale, $m_q = (m_u + m_d)/2$, and $d_{m_q} = (d_{m_u}m_u + d_{m_d}m_d)/(m_u + m_d)$.

With the variations of the fundamental constants, the fractional frequency ratio of two atomic clocks A and B changes such that

$$\frac{\delta(f_\mathrm{A}/f_\mathrm{B})}{f_\mathrm{A}/f_\mathrm{B}} = k_\alpha\frac{\delta\alpha}{\alpha} + k_{m_e}\frac{\delta(m_e/\Lambda_{\mathrm{QCD}})}{m_e/\Lambda_{\mathrm{QCD}}} + k_{m_q}\frac{\delta(m_q/\Lambda_{\mathrm{QCD}})}{m_q/\Lambda_{\mathrm{QCD}}}, \tag{3}$$

where k_α, k_{m_e}, and k_{m_q} are sensitivity coefficients [48]. For the Yb/Cs ratio, atomic and nuclear structure calculations yield $k_\alpha = -2.52$, $k_{m_e} = -1$, and $k_{m_q} = 0.046$ [49–51]. Note that $m_e/\Lambda_{\mathrm{QCD}}$ is used in Eq. (3) instead of the proton-to-electron mass ratio [52] in conventional formula used in previous atomic clock experiments [26–36]. From Eqs. (2) and (3), the relationship between $\delta(f_{\mathrm{Yb}}/f_{\mathrm{Cs}})/(f_{\mathrm{Yb}}/f_{\mathrm{Cs}})$ and φ is obtained. When the scalar field oscillates as $\varphi(t) \sim \varphi_0\cos(2\pi ft)$ with a frequency given by the Compton frequency $f = m_\varphi c^2/h$ (h, Planck constant), and carries an energy density of $\rho_\varphi = c^2\pi f^2\varphi_0^2/(2G)$ (G, Newtonian constant of gravitation) [43], the frequency ratio oscillates such that $\delta(f_{\mathrm{Yb}}/f_{\mathrm{Cs}})/(f_{\mathrm{Yb}}/f_{\mathrm{Cs}}) \sim A\cos(2\pi ft)$. Assuming that ρ_φ consists of the local dark matter density $\rho_{\mathrm{DM}} \sim 0.3$ GeV/cm^3, the relationship between the amplitude A and the coupling coefficients is obtained as

$$A = [-2.52d_\alpha - d_{m_e} + 0.046d_{m_q} + 0.954d_g]\sqrt{\frac{2G\rho_{\mathrm{DM}}}{\pi f^2c^2}}. \tag{4}$$

The goal of the analysis is to estimate the amount of A in the experimental data.

The experimental setup is schematically described in Fig. 1. At National Metrology Institute of Japan (NMIJ), we developed two atomic clocks: an Yb optical lattice clock (NMIJ-Yb1) and a Cs fountain clock (NMIJ-F2). The details of the experimental apparatuses of NMIJ-Yb1 and NMIJ-F2 are described in Refs. [45,53–55], and thus we here provide a brief description. NMIJ-Yb1 uses the ^{1}S$_0$-^{3}P$_0$ electronic transition of ^{171}Yb at 518 THz, and NMIJ-F2 the $|F=3, m_F=0\rangle \to |F=4, m_F=0\rangle$ hyperfine transition of the ground state of ^{133}Cs at 9.2 GHz. These two clocks are compared through a H maser that generates the Coordinated Universal Time of NMIJ [UTC (NMIJ)] with an auxiliary output generator (AOG). The frequency of NMIJ-Yb1 is compared with that of UTC (NMIJ) by counting a beat frequency between an ultrastable laser for probing the transition of Yb and an optical frequency comb [56] that is phase locked to UTC(NMIJ). NMIJ-Yb1 is then linked to the H maser with the AOG frequency measured by a time-interval counter. The frequency of NMIJ-F2 is compared with that of the H maser via an ultrastable cryogenic sapphire oscillator (CSO) [57]

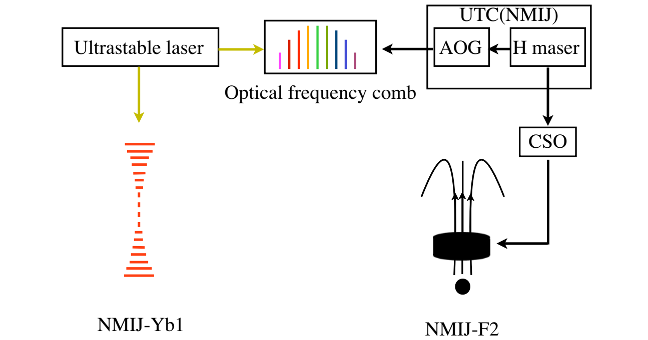

FIG. 1. Experimental setup. AOG, auxiliary output generator; CSO, cryogenic sapphire oscillator.

used as a local oscillator of NMIJ-F2. Both NMIJ-Yb1 and NMIJ-F2 are operated nearly continuously. The operation of NMIJ-Yb1 is supported by a reliable laser system based on a frequency comb [58], automatic laser relock schemes [59], and remote controlling systems. The systematic frequency shifts of NMIJ-Yb1 and NMIJ-F2 are compensated according to methods described in Refs. [45,53–55]. Some of the systematic shifts of NMIJ-Yb1 are reevaluated as described in the Supplemental Material [60].

During a search period of $T_{\text{total}} = 298$ days from November 17, 2020, to September 11, 2021, the measurement of the frequency ratio Yb/Cs was carried out in two campaigns with periods of (i) 25 days from November 17, 2020, and (ii) 40 days from August 2, 2021, with uptimes of 64.4% and 74.5%, respectively. Figure 2(a) shows the data points of the fractional frequency ratio. The data were averaged over an interval of 1×10^4 s, since this Letter aims to search for oscillations at low frequencies. The total number of the averaged data was $N_{\text{data}} = 447$. Figure 2(b) shows the Allan deviation of the ratio measurement, indicating the domination of the white frequency noise in the Yb/Cs data.

To estimate the strength of harmonic oscillation signals in the measured Yb/Cs ratio, we employed an analysis method similar to those of Refs. [24,41,43,82]. For each oscillation frequency f, we carried out the chi-square fit of the ratio data in Fig. 2(a) by a function $p_1 \cos(2\pi f t) + p_2 \sin(2\pi f t) + p_3$ with free parameters p_1, p_2, and p_3, and then obtained the amplitude as $A = \sqrt{p_1^2 + p_2^2}$. The analyzed frequencies were chosen from $1/T_{\text{total}} \sim 3.9 \times 10^{-8}$ to 4.9×10^{-5} Hz, corresponding to the mass range from $m_\varphi = 1.6 \times 10^{-22}$ to 2.0×10^{-19} eV/c^2. The frequency

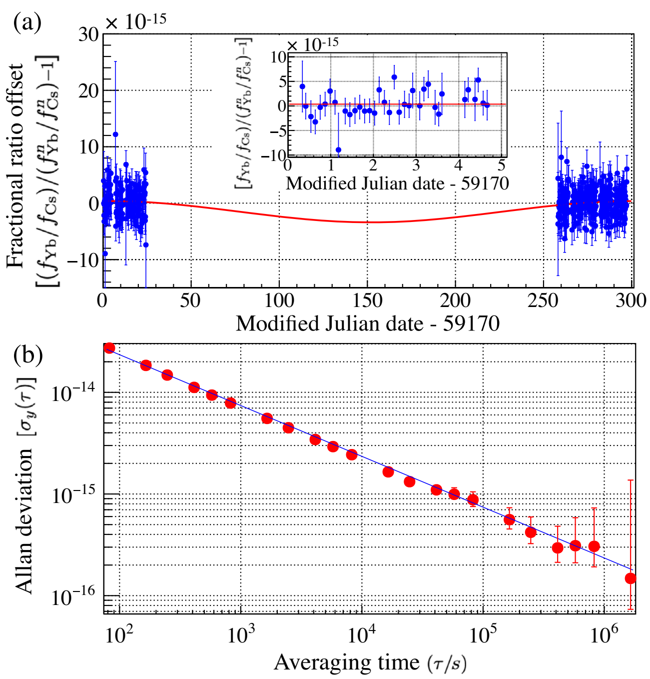

FIG. 2. (a) Fractional frequency ratio of Yb/Cs averaged over an interval of 1×10^4 s from modified Julian date 59170 (November 17, 2020). $(f_{\text{Yb}}/f_{\text{Cs}})/(f^n_{\text{Yb}}/f^n_{\text{Cs}}) - 1$ in the vertical axis denotes the fractional offset of the measured ratio from the ratio calculated by nominal frequencies $f^n_{\text{Cs}} = 9\,192\,631\,770$ Hz and $f^n_{\text{Yb}} = 518\,295\,836\,590\,863.6$ Hz [47]. The error bar indicates the combined statistical and systematic uncertainty of the frequency measurement [60], dominated by the statistical uncertainty. The red curve shows an example of the fit of a sinusoidal function. The inset shows an enlarged view of the data points. (b) Allan deviation of the frequency ratio calculated from concatenated data. The blue line indicates a slope of $2.3 \times 10^{-13}/\sqrt{(\tau/s)}$.

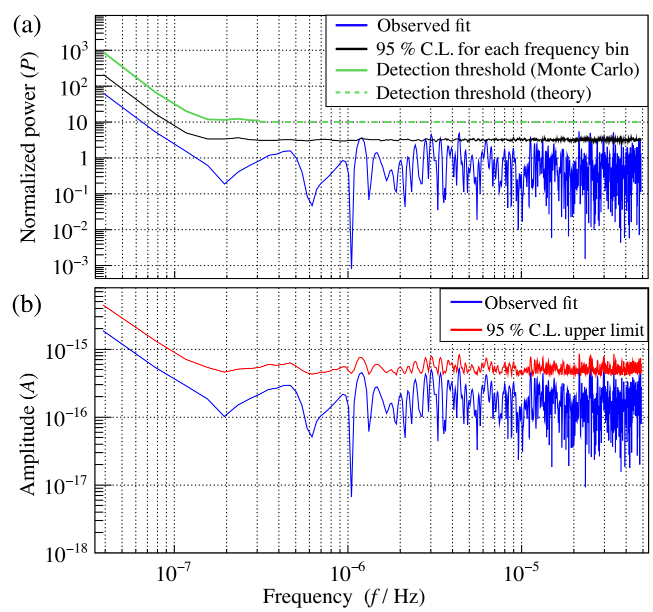

FIG. 3. (a) Normalized power spectrum $P = A^2 N_{\text{data}}/(4\sigma^2)$ obtained from the fit (blue line). The black line shows the 95% confidence limit (C.L.) of the simulated noise spectrum for each frequency bin. The green solid and dashed lines indicate the detection thresholds (see text) calculated by a Monte Carlo simulation and a theoretical calculation, respectively. (b) Amplitude spectrum A (blue line) and upper limits of A at the 95% confidence level (red line).

bin width was determined by $\Delta f = 1/T_{\mathrm{total}}$, resulting in the total bin number of $N_f = 1262$. We also defined the normalized power spectrum $P = A^2 N_{\mathrm{data}}/(4\sigma^2)$ [43,82], where $\sigma^2 = (2.5 \times 10^{-15})^2$ denotes the variance of the Yb/Cs data. Figures 3(a) and 3(b) show the obtained power P and amplitude A as a function of f, respectively (blue lines).

To determine whether we observed a signal exceeding a noise level, an expected noise power spectrum was calculated by a Monte Carlo simulation with a white frequency noise model [60]. The black line in Fig. 3(a) shows the 95% confidence level of the noise spectrum for each frequency bin. We found several events exceeding this level, since many frequency bins ($N_f = 1262$) were investigated. To take account of this so-called look-elsewhere effect, we defined a detection threshold [24,43] such that the probability of finding a noise above this threshold is 5% for a search involving N_f bins [83]. The detection threshold was calculated by a Monte Carlo simulation and a theoretical calculation [60], and is shown in Fig. 3(a) (green line). We observed no signals exceeding the detection threshold. Figure 3(b) shows upper limits of the amplitudes at the 95% C.L. (red line) [60]. These limits are model-independent results of this Letter.

Upper bounds on the coupling coefficients d_α, d_{m_e}, d_{m_q}, and d_g were transformed from the amplitude limits using Eq. (4), assuming that these respective couplings dominate. Interferences between the scalar field oscillations arise from a velocity distribution of the scalar fields in our Galaxy, resulting in a temporal variation of the field amplitude φ_0 [84]. Since search periods by atomic clocks are typically shorter than expected coherence times of the field oscillations (e.g., > 600 years in our target mass regions) [85], the atomic clock experiments only sample one of the possible values of φ_0, indicating that φ_0 should not be treated as deterministic but stochastic. With an assumed probability distribution of φ_0 [84], the 95% C.L. on d_α, d_{m_e}, d_{m_q}, and d_g was therefore rescaled by a factor of 3.0.

Figures 4(a)–4(d) show our 95% C.L. on the coupling coefficients as a function of m_φ, together with those derived from previous atomic clock measurements [41–43], an optical clock network [24], Dy spectroscopy [82], and also equivalence principle tests in which differential

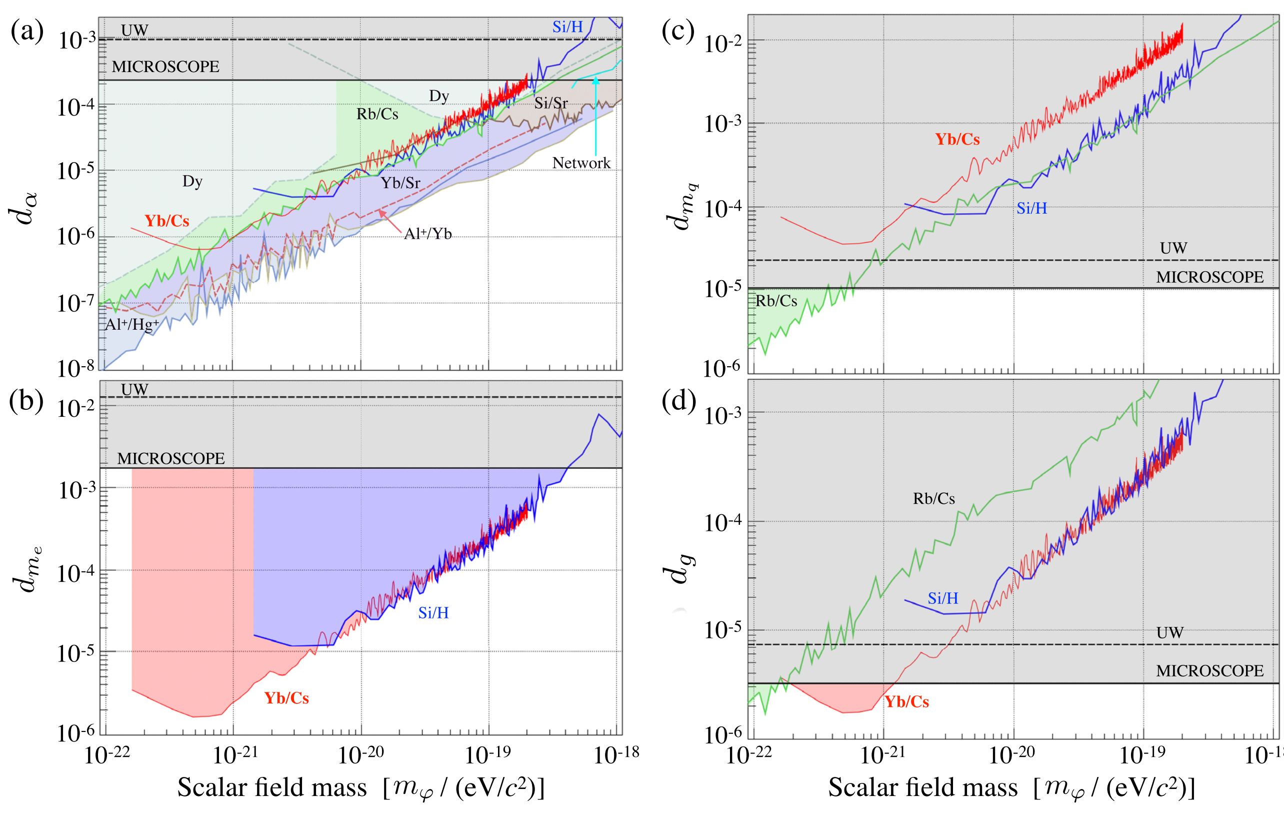

FIG. 4. Exclusion plots for (a) d_α, (b) d_{m_e}, (c) d_{m_q}, and (d) d_g at the 95% confidence level, assuming that these respective couplings dominate. The shaded areas show excluded regions set by our Yb/Cs measurement, previous atomic clock measurements of Si/Sr [41], Yb/Sr [42], $\mathrm{Al^+/Hg^+}$ [42], $\mathrm{Al^+/Yb}$ [42], Rb/Cs [43], Si/H [41] ratios, an optical clock network [24], Dy spectroscopy [82], equivalence principle tests by University of Washington (UW) [86], and the MICROSCOPE satellite experiment [87]. To take account of interferences between the scalar field oscillations [84], the limits derived from the atomic clocks and the Dy spectroscopy are rescaled by a factor of 3.0, which reduces the exclusion areas.

accelerations between two macroscopic objects have been measured [86,87]. The equivalence principle tests search for a Yukawa force mediated by the virtual exchange of the scalar field. Thus, the limits set by these tests are not affected by the interference effect of the oscillation. We set new limits on d_{m_e} at the range from $m_\varphi = 10^{-22}$ to 10^{-20} eV$/c^2$. The most stringent limit was $d_{m_e} \lesssim 1.6 \times 10^{-6}$ at $m_\varphi \sim 5 \times 10^{-22}$ eV$/c^2$, which was improved by a factor of $\sim 10^3$. In spite of the relatively stringent constraints on d_g by the equivalence principle tests, which is because the mass of a macroscopic object mostly consists of the nucleon mass [18,19], we improved the limits on d_g at the range from $m_\varphi = 10^{-22}$ to 10^{-21} eV$/c^2$. While the constraints on the other coefficients d_α and d_{m_q} are not improved in this Letter, our limits are complementary to those of previous works.

Since the frequency of NMIJ-F2 realizes the definition of the SI second, the measured Yb/Cs ratio yields the absolute frequency of Yb. From the weighted mean value of the ratio in Fig. 2(a), we obtained the absolute frequency $f_{\rm Yb} = 518\,295\,836\,590\,863.69(28)$ Hz with a fractional uncertainty of 5.3×10^{-16} [60]. Our measured frequency was in good agreement with previous values [34,45,88–91] at the 10^{-16} levels. We note that the previous values were mostly obtained by remote comparisons with some specific Cs fountains via satellite links, implying that correlations exist among these values. Our local Yb/Cs measurement provides an independent result with its uncertainty better than those of previous local measurements [88,92].

In conclusion, we have searched for harmonic oscillation signals from long-term comparison data between NMIJ-Yb1 and NMIJ-F2. We improved constraints on d_{m_e} in the mass range from $m_\varphi = 10^{-22}$ to 10^{-20} eV$/c^2$ and d_g in the range of $m_\varphi \lesssim 10^{-21}$ eV$/c^2$, assuming that these coupling strengths dominate. Especially, limits on d_{m_e} at $m_\varphi \sim 5 \times 10^{-22}$ eV$/c^2$ were improved by 3 orders of magnitude. This Letter has demonstrated that long-term operation of an optical clock extends the discovery reach of dark matter searches, which can motivate other groups to extend operation periods [93]. Robust optical clocks should also contribute to the search for annual variations of fundamental constants [36] and topological defects [24,25].

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Note added.—While completing the manuscript, we found recent results of the MICROSCOPE experiment that have improved the limit on the violation of the equivalence principle by a factor of 4.6 [94].

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