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1	Resolving the gravitational redshift within a millimeter atomic
2	sample
3 4	Tobias Bothwell ^{1,*} , Colin J. Kennedy ^{1,2} , Alexander Aeppli ¹ , Dhruv Kedar ¹ , John M. Robinson ¹ , Eric Oelker ^{1,3} , Alexander Staron ¹ , and Jun Ye ^{1,*}
5	¹ JILA, National Institute of Standards and Technology and University of Colorado
6	Department of Physics, University of Colorado
7	Boulder, Colorado 80309-0440, USA
8	² Present Address: Honeywell Quantum Solutions, Broomfield, Colorado 80021
9	³ Present Address: Physics Department, University of Glasgow
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Einstein's theory of general relativity states that clocks at different gravitational 11 potentials tick at different rates – an effect known as the gravitational redshift¹. As 12 fundamental probes of space and time, atomic clocks have long served to test this 13 prediction at distance scales from 30 centimeters to thousands of kilometers²⁻⁴. 14 Ultimately, clocks will study the union of general relativity and quantum mechanics 15 once they become sensitive to the finite wavefunction of quantum objects oscillating in 16 curved spacetime. Towards this regime, we measure a linear frequency gradient 17 18 consistent with the gravitational redshift within a single millimeter scale sample of ultracold strontium. Our result is enabled by improving the fractional frequency 19 measurement uncertainty by more than a factor of 10, now reaching 7.6×10^{-21} . This 20 21 heralds a new regime of clock operation necessitating intra-sample corrections for gravitational perturbations. 22

Modern atomic clocks embody Arthur Schawlow's motto to "never measure anything but frequency." This deceptively simple principle, fueled by the innovative development of laser science and quantum technologies based on ultracold matter, has led to dramatic progress in clock performance. Recently, clock measurement precision reached the mid-19th digit in one

hour^{5,6}, and three atomic species achieved systematic uncertainties corresponding to an error
equivalent to less than 1 s over the lifetime of the Universe⁷⁻¹⁰. Central to this success in
neutral atom clocks is the ability to maintain extended quantum coherence times while using
large ensembles of atoms^{5,6,11}. The pace of progress has yet to slow. Continued improvement
in measurement precision and accuracy arising from the confluence of metrology and quantum
information science¹²⁻¹⁵ promises discoveries in fundamental physics¹⁶⁻²⁰.

Clocks fundamentally connect space and time, providing exquisite tests of the theory of 33 general relativity. Hafele and Keating took cesium-beam atomic clocks aboard commercial 34 airliners in 1971, observing differences between flight-based and ground-based clocks 35 consistent with special and general relativity²¹. More recently, RIKEN researchers compared 36 two strontium optical lattice clocks (OLCs) separated by 450 m in the Tokyo Skytree, resulting 37 in the most precise terrestrial redshift measurement to date²². Proposed satellite-based 38 measurements^{23,24} will provide orders of magnitude improvement to current bounds on 39 gravitational redshifts^{3,4}. Concurrently, clocks are anticipated to begin playing important roles 40 for relativistic geodesy²⁵. In 2010 Chou *et al.*² demonstrated the precision of their Al⁺ clocks by 41 measuring the gravitational redshift resulting from lifting one clock vertically by 30 cm in 40 42 hours. With a decade of advancements, today's leading clocks are poised to enable local 43 geodetic surveys of elevation at the sub-centimeter level on Earth, complimenting spatial 44 averaging techniques²⁶. 45

Atomic clocks strive to simultaneously optimize measurement precision and systematic uncertainty. For traditional OLCs operated with one-dimensional (1D) optical lattices, achieving low instability has involved the use of high atom numbers at trap depths sufficiently large to suppress tunnelling between neighboring lattice sites. While impressive performance has been

achieved, effects arising from atomic interactions and AC Stark shifts associated with the 50 trapping light challenge advancements in OLCs. Here we report a new operational regime for 51 1D OLCs, both resolving the gravitational redshift across our atomic sample and 52 synchronously measuring a fractional frequency uncertainty of 7.6×10⁻²¹ between two 53 uncorrelated regions. Our system employs ~100,000 ⁸⁷Sr atoms at ~100 nK loaded into a 54 shallow, large waist optical lattice, reducing both AC Stark and density shifts. Motivated by our 55 earlier work on spin-orbit coupled lattice clocks^{27,28}, we engineer atomic interactions by 56 operating at a 'magic' trap depth, effectively removing collisional frequency shifts. These 57 advances enable record optical atomic coherence (37 s) and expected single clock stability 58 (3.1×10⁻¹⁸ at 1 s) using macroscopic samples, paving the way toward lifetime limited OLC 59 operation. 60

Central to our experiment is an in-vacuum optical cavity (Fig. 1a and Methods) for 61 power enhancement of the optical lattice. The cavity (finesse 1300) ensures wavefront 62 homogeneity of our 1D lattice while the large beam waist (260 µm) reduces the atomic density 63 by an order of magnitude compared to our previous system¹⁰. We begin each experiment by 64 trapping fermionic ⁸⁷Sr atoms into the 1D lattice at a trap depth of 300 lattice photon recoil 65 energies (E_{rec}), loading a millimeter scale atomic sample (Fig. 1a). Atoms are simultaneously 66 cooled and polarized into a single nuclear spin before the lattice is adiabatically ramped to an 67 operational depth of 12 E_{rec}. Clock interrogation proceeds by probing the ultranarrow ${}^{1}S_{0}(q) \rightarrow$ 68 ${}^{3}P_{0}(e)$ transition with the resulting excitation fraction measured by fluorescence spectroscopy. 69 Scattered photons are collected on a camera, enabling in-situ measurement with 6 µm 70 71 resolution, corresponding to ~15 lattice sites.

Quantum state control has been vital to recent advances in atom-atom and atom-light 72 coherence times in 3D OLCs and tweezer clocks^{5,11,15}. Improved quantum state control is 73 demonstrated through precision spectroscopy of the Wannier-Stark states of the OLC^{29,30}. The 74 1D lattice oriented along gravity has the degeneracy of neighboring lattice sites lifted by the 75 gravitational potential energy. In the limit of shallow lattice depths, this creates a set of 76 delocalized states. By ramping the lattice depth to 6 E_{rec}, much lower than in traditional 1D 77 lattice operations^{7,8,10}, clock spectroscopy probes this delocalization (Fig. 1d). The ability to 78 engineer the extent of atomic wavefunctions through the adjustment of trap depth creates an 79 opportunity to control the balance of on-site *p*-wave versus neighboring-site *s*-wave atomic 80 interactions. We utilize this tunability by operating at a 'magic' trap depth³¹, where the 81 frequency shifts arising from on-site and off-site atomic interactions cancel, enabling a 82 reduction of the collisional frequency shifts by more than three orders of magnitude compared 83 our previous work¹⁰. 84

Extended atomic coherence times are critical for both accuracy and precision. An 85 aspirational milestone for clock measurement precision is the ability to coherently interrogate 86 atomic samples up to the excited state's natural lifetime. To evaluate the limits of our clock's 87 88 atomic coherence, we perform Ramsey spectroscopy to measure the decay of fringe contrast as a function of the free-evolution time. By comparing two uncorrelated regions within our 89 atomic sample, we determine the contrast and relative phase difference between the two sub-90 ensembles (Fig. 2). The contrast decays exponentially with a time constant of 37 s (quality 91 factor of 3.6×10^{16}), corresponding to an additional decoherence time of 53 s relative to the ${}^{3}P_{0}$ 92 natural lifetime (118 s)³². This represents the longest optical atomic coherence time measured 93 in any spectroscopy system to date. 94

We utilize Rabi spectroscopy in conjunction with *in-situ* imaging to microscopically 95 probe clock transition frequencies along the entire vertically oriented atomic ensemble. With a 96 standard interleaved probing sequence using the $|g, m_F = \pm \frac{5}{2}$ to $|e, m_F = \pm \frac{3}{2}$ transitions for 97 minimal magnetic sensitivity, we reject the first order Zeeman shifts and vector AC Stark shifts. 98 The *in-situ* imaging of atoms in the lattice allows measurement of unprocessed frequencies 99 across the entire atomic sample (Fig. 1a and Methods). The dominant differential perturbations 100 arise from atom-atom interactions (residual density shift contributions after we operate at the 101 'magic' trap depth) and magnetic field gradients giving rise to pixel-specific 2nd order Zeeman 102 shifts. Using the total camera counts and m_F-dependent frequency splitting, we correct the 103 density and 2nd order Zeeman shift at each pixel. These corrections result in the processed 104 105 frequencies per pixel shown in Fig. 3a, with error bars representing the quadrature sum of statistical uncertainties from the center frequency, the density shift correction, and the 2nd order 106 Zeeman shift correction. Additional systematics are described in the Methods. This approach 107 demonstrates an efficient method for rapid and accurate evaluation of various systematic 108 109 effects throughout a single atomic ensemble. Unlike traditional 1D OLCs where systematic 110 uncertainties are quoted as global parameters, we now microscopically characterize these effects. 111

112 This new microscopic *in-situ* imaging allows determination of the gravitational redshift 113 within a single atomic sample, probing an uncharacterized fundamental clock systematic. Two 114 identical clocks on the surface of a planet separated by a vertical distance *h* will differ in 115 frequency (δf) as given by

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$$\frac{\delta f}{f} = \frac{ah}{c^2} \quad (Eq. 1),$$

with *f* the clock frequency, *c* the speed of light, and *a* the gravitational acceleration. The gravitational redshift at Earth's surface corresponds to a fractional frequency gradient of $-1.09 \times$ 10^{-19} /mm in the coordinate system of Fig. 1a. Measurement of a vertical gradient across the atomic sample consistent with the gravitational redshift provides an exquisite verification of an individual atomic clock's frequency control.

122 Our intra-cloud frequency map (Fig. 3a) allows us to evaluate gradients across the atomic sample. Over 10 days we performed 14 measurements (ranging in duration from 1-17 123 hours) to search for the gravitational redshift across our sample. For each we fit a linear slope 124 and offset after taking into account density shift and 2nd order Zeeman corrections, reporting 125 the slope in Fig. 3b. From this measurement campaign we find the weighted mean (standard 126 error of the weighted mean) of the frequency gradient in our system to be $-1.00(12) \times 10^{-19}$ /mm. 127 We evaluate additional differential systematics (see Methods) and find a final frequency 128 gradient of $-9.8(2.3) \times 10^{-20}$ /mm, consistent with the predicted redshift. 129

The ability to resolve the gravitational redshift within our system suggests a level of 130 frequency control beyond previous clock demonstrations, vital for the continued advancement 131 of clock accuracy and precision. Previous fractional frequency comparisons¹⁵ have reached 132 uncertainties as low as 4.2×10⁻¹⁹. Similarly, we perform a synchronous comparison between 133 two uncorrelated regions of our atomic cloud (Fig. 4a). By binning ~100 pixels per region, we 134 substantially reduce instability caused by quantum projection noise³³. Analyzing the frequency 135 difference between regions from 92 hours of data, we find a fractional frequency instability of 136 4.4×10⁻¹⁸/ $\sqrt{\tau}$ (τ is the averaging time in seconds), resulting in a fractional frequency uncertainty 137 of 7.6×10⁻²¹ for full measurement time, nearly two orders of magnitude lower than the previous 138 record. From this measurement we infer a single region instability of $3.1 \times 10^{-18} / \sqrt{\tau}$. Dividing the 139

fractional frequency difference by the spatial separation between each region's center of mass gives a frequency gradient of $-1.30(18)\times10^{-19}$ /mm. Correcting for additional systematics as before results in a gradient of $-1.28(27)\times10^{-19}$ /mm, again fully consistent with the predicted redshift.

In conclusion, we have established a new paradigm for atomic clocks. The vastly 144 145 improved atomic coherence and frequency homogeneity throughout our sample allow us to resolve the gravitational redshift at the submillimeter scale, observing for the first time the 146 frequency gradient from gravity within a single sample. We demonstrate a synchronous clock 147 comparison between two uncorrelated regions with a fractional frequency uncertainty of 148 7.6×10⁻²¹, advancing precision by nearly two orders of magnitude. These results suggest that 149 there are no fundamental limitations to inter-clock comparisons reaching frequency 150 uncertainties at the 10⁻²¹ level, offering new opportunities for tests of fundamental physics. 151

Fig. 1: Experimental system and quantum state control. a, A millimeter length sample of 153 ~100,000 ⁸⁷Sr atoms are trapped in a 1D optical lattice formed within an in-vacuum cavity. The 154 longitudinal axis of the cavity, z, is oriented along gravity. We probe atoms along the ${}^{1}S_{0} \rightarrow {}^{3}P_{0}$ 155 transition using a clock laser locked to an ultrastable crystalline silicon cavity^{6,34}. **b**, Rabi 156 spectroscopy with a 3.1 s pulse time. Open purple circles indicate data with a corresponding 157 Rabi fit in green. **c**, Neighboring lattice sites are detuned by gravity, creating a Wannier-Stark 158 ladder. Clock spectroscopy probes the overlap of Wannier-Stark states between lattice sites 159 that are m sites away with Rabi frequency Ω_m . d, Rabi spectroscopy probes Wannier-Stark 160 state transitions, revealing wavefunction delocalization of up to 5 lattice sites. The number of 161 162 lattice sites is indicated above each transition, with blue(red) denoting Wannier-Stark transitions to higher(lower) lattice sites. 163

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Fig. 2: Atomic coherence. We use Ramsey spectroscopy with a randomly sampled phase for 166 the second pulse to determine the coherence time of our system¹¹. a, We measure the 167 excitation fraction across the cloud, shown in purple for a single measurement, and calculate 168 the average excitation fractions in regions p_1 and p_2 , separated by 2 pixels. **b**, Parametric plots 169 of the excitation fraction of p_1 versus p_2 in purple for 6 s, 30 s and 50 s dark time demonstrate 170 171 a phase shift between the two regions and contrast decay. Using a maximum likelihood estimator, we extract the phase and contrast for each dark time with the fit, shown in green. c, 172 Contrast decay as a function of time in green is fit with an exponential decay in gold, giving an 173 atomic coherence decay time of 36.5(0.7) s and a corresponding quality factor of 3.6×10^{16} . 174 175 After accounting for the finite radiative decay contribution, we infer an additional decoherence time constant of 52.8(1.5) s. 176

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Figure 3: Evaluating frequency gradients. a, For each measurement we construct a 180 microscopic frequency map across the sample, with raw frequencies shown in green. The 2nd 181 order Zeeman correction is shown as a dashed gold line. Processed frequencies shown in 182 purple include both density shift corrections and 2nd order Zeeman corrections, with 183 uncertainties arising from the quadrature sum of statistical, density shift correction, and 2nd 184 order Zeeman correction uncertainties. To this we fit a linear function, shown in black. **b**, Over 185 the course of 10 days, we completed 14 measurements. For each measurement, we create a 186 corrected frequency map and fit a linear slope as in **a**. This slope is plotted for each 187 measurement, as well as a weighted mean (black) with associated statistical uncertainty 188 189 (dashed black) and total uncertainty as reported in Table 1 (dotted black). The expected gravitational gradient is shown in red. All data is taken with Rabi spectroscopy using a 3.1 s π-190 191 pulse time except for 08/13 which used a 3.0 s pulse time. The reduced chi-square statistic is 3.0, indicating a small underestimation of error variances entirely consistent with the additional 192 systematic uncertainties in Table 1. 193

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Figure 4: *In-situ* synchronous clock comparison. **a**, The cloud is separated as in Fig. 2a. The gravitational redshift leads to the higher clock(blue) ticking faster than the lower one(red). The length scale is in millimeters. **b**, Allan deviation of the frequency difference between the two regions in **a** over 92 hours. Purple points show fractional frequency instability fit by the solid green line, with the quantum projection noise limit indicated by the dashed black line. We attribute the excess instability of the measurement relative to QPN to detection noise. The expected single atomic region instability is shown in gold.

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Methods

211 In-Vacuum Cavity

Central to our system is an in-vacuum lattice buildup cavity oriented along gravity (Fig. 212 1a). Two mirrors with radius of curvature of 1 m are separated by ~15 cm, achieving a mode 213 waist of 260 µm. Our over-coupled cavity has a finesse at the lattice wavelength (813 nm) of 214 ~1300 and a power buildup factor of ~700 (ratio of circulating to input intensity). This enables 215 lattice depths in excess of 500 E_{rec} (lattice photon recoil energy) using a diode-based laser 216 system. The dimensional stability of the cavity combined with the simplified diode laser system 217 enables robust operation compared with our previous Ti:Sapphire retro-reflected design¹⁰. The 218 cavity mirrors are anti-reflection coated at the clock wavelength of 698 nm. 219

220 One cavity mirror is mounted to a piezo for length stabilization while the other mirror is 221 rigidly mounted for phase reference for the clock laser. Grounded copper shields between 222 atoms and mirrors prevent DC Stark induced shifts due to charge buildup on the mirrors and 223 piezo^{35,36}. Each shield (5 mm thick) has a centered hole of 6 mm diameter to accommodate 224 the optical lattice beam, with shielding performance verified through evaluation of the DC Stark 225 shift systematic.

226 Atomic sample preparation

⁸⁷Sr atoms are cooled and loaded into a 300 E_{rec} optical lattice using standard two stage magneto-optical trapping techniques¹⁰. Once trapped, atoms are simultaneously nuclear spin polarized, axially sideband cooled, and radially doppler cooled into a single nuclear spin state at temperatures of 800 nK. The lattice is then adiabatically ramped to the operational trap depth of 12 E_{rec} , where a series of pulses addressing the clock transition prepares atoms into

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 $|g, m_F = \pm \frac{5}{2}\rangle$. Clock spectroscopy is performed by interrogating the $|g, m_F = \pm \frac{5}{2}\rangle$ to $|e, m_F = \pm \frac{3}{2}\rangle$ transition, the most magnetically insensitive ⁸⁷Sr clock transition⁶.

234 **Imaging**

The clock excitation fraction is read out using standard fluorescence spectroscopy techniques^{6,10,37}. Photons are collected on both a photo-multiplier tube for global readout and electron multiplying charge coupled device camera for an *in-situ* readout of clock frequency. Camera readout is performed in full vertical binning mode, averaging the radial dimension of the lattice. This provides 1D *in-situ* imaging for all synchronous evaluations.

We use a 25 μ s fluorescence probe with an intensity of I/I_{sat} ~ 20 (I_{sat} being the saturation intensity), ensuring uniform scattering across the atomic sample. Before imaging, the optical lattice is ramped back to 300 E_{rec} to decrease imaging aberration resulting from the extended radial dimension at 12 E_{rec}.

244 Analysis

Standard clock lock techniques and analysis are used^{6,11,33}, with differences in 245 excitation fraction converted to frequency differences using Rabi lineshapes. Each dataset is 246 composed of a series of clock locks, tracking the center of mass frequency of the atomic 247 sample. A clock lock is four measurements probing alternating sides of the Rabi lineshape for 248 opposite nuclear spin transitions. Frequency corrections based on excitation fraction become 249 ambiguous when the excitation fraction measured is consistent with the Rabi lineshape at 250 multiple detunings. To avoid erroneous frequency corrections, we remove clock locks with 251 excitation fractions above (below) .903×C (.116×C), where C is the Rabi contrast. From each 252

clock lock, a pixel specific center frequency f_i and frequency splitting between opposite m_F states Δ_i are calculated, creating an *in-situ* frequency map of the 1D atomic sample. This allows rejection of vector shifts on a pixel-by-pixel basis and probes the magnetic field induced splitting of m_F transitions. The atom weighted mean frequency is subtracted from every lock cycle to reject common mode laser noise.

258 For each dataset we approximate the atomic profile with a Gaussian fit, identifying a 259 center pixel and associated Gaussian width (σ). All analysis is performed within the central region of $\pm 1.5\sigma$ which demonstrates the lowest frequency instability. Identifying a center pixel 260 261 for data processing ensures rejection of any day-to-day drift in the position of the cloud due to varying magnetic fields modifying MOT operation on the narrow line transition. The density 262 shift coefficient (see Density Shift section) is derived from the average center frequency per 263 pixel. Using this coefficient, we correct f_i and Δ_i for the density shift. 2nd order Zeeman 264 corrections using these updated frequencies are then applied. 265

Gradient analysis is based on the processed center frequencies per pixel. A linear fit to the frequencies as a function of pixel is performed using least squares, with uncertainty per pixel arising from the quadrature sum of statistical frequency uncertainty, statistical 2nd order Zeeman uncertainty, and density shift correction uncertainty.

For the two-clock comparison (Fig. 4b), all data from 8/14-8/22 was taken with the same duty cycle and π -pulse time (3.1 s). Data was processed relative to a fit center pixel as discussed and concatenated. Two equal regions extend from the center of the sample to a width of ±1.5 σ , with two empty pixels between regions to ensure uncorrelated samples. Each region is processed for the atom weighted mean frequency, enabling a synchronous frequency comparison between two independent clocks.

276 Atomic Coherence

We use a Ramsey sequence to measure the atomic coherence. We prepare a sample in the $|g, m_F = +\frac{5}{2}\rangle$ state and apply a $\pi/2$ pulse along the $|g, m_F = +\frac{5}{2}\rangle$ to $|e, m_F = +\frac{3}{2}\rangle$ transition. After waiting for a variable dark time, we apply a second $\pi/2$ pulse with a random phase relative to the first. We then measure the excitation fraction.

Two regions, p₁ and p₂, are identified using the same technique as in the synchronous 281 instability measurement. For each experimental sequence, we find the average excitation 282 fraction in p₁ and p₂. A mean frequency shift across the sample primarily due to a magnetic 283 field gradient creates a differential phase as a function of time between p_1 and p_2 . We create a 284 parametric plot of the average excitation in p1 and p2 for each dark time and use a maximum 285 likelihood estimator to fit an ellipse to each dataset, calculating phase and contrast^{15,38}. To 286 estimate uncertainty in the contrast for each dark time a bootstrapping technique is used¹¹. 287 Fitting the contrast as a function of dark time with a single exponential returns an effective 288 atomic coherence time. 289

290 Systematics

291 **Imaging**

We calibrate our pixel size using standard time of flight methods: we observe an atomic sample in freefall for varying times to determine an effective pixel size along the direction of gravity. Immediately after our 10-day data campaign we measured our effective pixel size to be $6.04 \mu m$. Due to thermal drift of our system, the pixel size can vary by up to 0.5 μm /pixel over months which we take as the calibration uncertainty. Spatial correlations may limit imaging resolution. We measure these correlations by
 placing atoms into a superposition of clock states. Any measured spatial correlation is due to
 the imaging procedure. In our system we find no correlations between neighboring pixels¹¹.
 The optical resolution of our imaging lens is specified at 2 µm.

Lattice tilt from gravity will modify the measured gradient. We find the lattice tilt in the imaging plane to be 0.11(0.06) degrees, providing an uncertainty orders of magnitude smaller than the pixel size uncertainty. We are insensitive to lattice tilt out of the imaging plane.

304 Zeeman Shifts

First order Zeeman shifts are rejected by probing opposite nuclear spin states⁶. The 2nd order Zeeman shift is given by

$$\Delta \nu_{B,2} = \xi \left(\Delta \nu_{B,1} \right)^2,$$

where $\Delta v_{B,1}$ is the splitting between opposite spin states and ξ the corresponding 2nd order Zeeman shift coefficient. For stretched spin state operation $(m_F = \pm \frac{9}{2})$, ξ =-2.456(3)×10⁻⁷ Hz⁻¹. Using known atomic coefficients³⁹ we find the 2nd order Zeeman coefficient for the $|g, m_F = \pm \frac{5}{2}\rangle$ to $|e, m_F = \pm \frac{3}{2}\rangle$ transition to be ξ_{op} =-1.23(8)×10⁻⁴ Hz, with the uncertainty arising from limited knowledge of atomic coefficients.

The 2nd Order Zeeman corrections are made for every clock lock (analogous to the insitu density shift corrections). For a typical day (8/13) the average 2nd order Zeeman gradient is -7.0×10^{-20} /mm, corresponding to a splitting between opposite nuclear spin states of 12.7 mHz/mm (0.291 mG/mm). We include an error of 4×10^{-21} /mm in Table 1 to account for the atomic uncertainty in the shift coefficient.

317 DC Stark

Electric fields perturb the clock frequency via the DC Stark effect. We evaluate gradients arising from this shift by using in-vacuum quadrant electrodes to apply bias electric fields in all three dimensions. We find a DC Stark gradient of $3(2)\times10^{-21}$ /mm.

321 Black Body Radiation Shift

The dominant frequency perturbation to room temperature neutral atom clocks is black body radiation (BBR). Similar to our previous work¹⁰, we homogenize this shift by carefully controlling the thermal surroundings of our vacuum chamber. Attached to the vacuum chamber are additional temperature control loops, with each vacuum viewport having a dedicated temperature control system. This ensures our dominant BBR contribution – high emissivity glass viewports – are all the same temperature to within 100 mK.

To bound possible BBR gradients, we introduce a 1 K gradient between the top and bottom of the chamber along the cavity axis by raising either the top or bottom viewports by 1 K. We compare these two cases and find no statistically significant changes in the frequency gradient across the entire sample. Accounting for uncertainty in linear frequency fits for each case, we estimate an uncertainty of 3×10^{-21} /mm. This finding is supported with a basic thermal model of the vacuum chamber.

334 **Density Shift**

Atomic interactions during Rabi spectroscopy lead to clock frequency shifts as a function of atomic density⁴⁰. For each gradient measurement, we evaluate the density shift coefficient χ_{dens} by fitting the average frequency *f* per pixel versus average camera counts per pixel *N* to an equation of the form

$$f(N) = \chi_{dens}N + B$$

Here B is an arbitrary offset. Once χ_{dens} is known, we remove the density shift at each pixel.

Residual density shift corrections may lead to error in our linear gradient. To bound this effect, we compare the density shift coefficient and gradient from our data run with a separate dataset at 8 E_{rec}. With the trap depth at 8 E_{rec} we found a linear gradient of s=-1.08×10⁻¹⁸/mm and a density shift coefficient of χ_8 =-1.39×10⁻⁶ Hz/count. During our data run we had an average density shift coefficient of χ_{op} =-2.43×10⁻⁸ Hz/count. We bound the uncertainty in our gradient from density shift as $\sigma_{den,unc} = |s \times \frac{\chi_{op}}{\chi_8}|$ =1.7×10⁻²⁰/mm.

347 Lattice Light Shifts

Lattice light shifts arise from differential AC Stark shifts between the ground and excited clock states. An approximate microscopic model of the lattice light shift (v_{LS}) in our system is given by⁴¹

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$$hv_{LS}(u,\delta_L) \approx \left(\frac{\delta\Delta\alpha^{E_1}}{\delta\nu}\delta_L - \Delta\alpha_{QM}\right)\frac{u^{\frac{1}{2}}}{2} - \left[\frac{\delta\alpha^{E_1}}{\delta\nu}\delta_L\right]u,$$

where *u* is the trap depth in units of E_{rec} , $\Delta \alpha^{E1}$ the differential electric dipole polarizability, $\Delta \alpha^{QM}$ the differential multi-polarizability, and $\delta_L = (\nu_L - \nu^{E1})$ the detuning between lattice frequency ν_L and effective magic frequency ν^{E1} . Our model has no dependence on the longitudinal vibrational quanta since we are in the ground vibrational band. We neglect higher order corrections from hyperpolarizability due to our operation at depths <60 E_{rec}. At our temperatures thermal averaging of the trap depth is a higher order correction (<5%) that is also neglected.

We model the linear differential lattice light shift across the atomic cloud as

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$$\frac{\delta h \nu_{LS}(u,\delta_L)}{\delta z} \approx \left[\frac{\left(\frac{\delta \alpha^{E_1}}{\delta \nu} \delta_L - \alpha_{QM} \right)}{4u^{1/2}} - \frac{\delta \alpha^{E_1}}{\delta \nu} \delta_L \right] \frac{\delta u}{\delta z}$$

where z is the coordinate corresponding to the axis of the cavity along gravity. To evaluate our differential lattice light shift at our operational depth we need δ_L and $\frac{\delta u}{\delta z}$. We modulate our lattice between two trap depths (u₁=14 E_{rec}, u₂=56 E_{rec}) and find our detuning from scalar magic frequency to be δ_L =7.4(0.6) MHz. To evaluate $\frac{\delta u}{\delta z}$ at our operational depth (u_{op}) we measure the linear gradient across the atomic cloud at δ_L + 250 MHz and δ_L - 250 MHz, the difference given by

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$$\frac{\delta h \nu_{LS}(u_{op}, \delta_{L+250 \text{ MHz}})}{\delta z} - \frac{\delta h \nu_{LS}(u_{op}, \delta_{L-250 \text{ MHz}})}{\delta z} \approx \left| \frac{1}{4u_{op}^{\frac{1}{2}}} - 1 \right| \frac{\delta \alpha^{E_1}}{\delta \nu} \delta_{500} \frac{\delta u_{op}}{\delta z},$$

where δ_{500} = 500 MHz. We find $\frac{\delta u_{op}}{\delta z}$ =0.0383/mm, which when combined with δ_L =7.4 MHz, gives us a fractional frequency gradient of -5×10⁻²¹/mm. Accounting for error in our lattice detuning and linear gradient gives us an uncertainty of 1×10⁻²¹/mm.

371 Other Systematics

For a 3.1 s π -pulse the probe AC Stark shift⁷ is -3(2)×10⁻²¹. A frequency scan of the $|g, m_F = -\frac{5}{2}\rangle$ to $|e, m_F = -\frac{3}{2}\rangle$ transition limits the variation of excitation fraction across the atomic sample to 1% or below, bounding any possible probe AC Stark gradient across the sample to (1×10^{-22}) .

376 Known Redshift

The gravitational acceleration (rounding to 4 digits) within our lab was evaluated by a USGS survey⁴² to be a=-9.796 m/s².

379 Systematic Budget

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Systematic	Slope (10 ⁻²⁰ /mm)	Uncertainty (10 ⁻²⁰ /mm)
Gradient (Fig. 3)	-10.0	1.2
BBR	0	0.3
Density	-	1.7
Lattice light shift	-0.5	0.1
DC Stark	0.3	0.2
Pixel Calibration	0	.8
2 nd Order Zeeman	-	.4
Other	0	<.1
Corrected Gradient	-9.8	2.3
Known Redshift	-10.9	<.1

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Table 1: Gradient Systematic Budget. Fractional frequency gradients and corresponding uncertainties. Fractional frequencies denoted with '-' are corrected on a pixel-by-pixel basis during initial data processing (Fig. 3a). The corrected gradient has known systematics removed with uncertainty given by the quadrature sum of all correction uncertainties.

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Author Contributions 399

All authors contributed to carrying out the experiments, interpreting the results, and writing the 400 manuscript. 401

*tobias.bothwell@colorado.edu, ye@jila.colorado.edu 402

Competing interests 403

The authors declare no competing interests. 404

Data and Code Availability 405

The experimental data and code analysis are available from the corresponding authors upon 406 reasonable request. 407

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409 **References**

- Einstein, A. Grundgedanken der allgemeinen Relativitätstheorie und Anwendung dieser Theorie
 in der Astronomie. *Preuss. Akad. der Wissenschaften, Sitzungsberichte* 315, (1915).
- 2. Chou, C. W., Hume, D. B., Rosenband, T. & Wineland, D. J. Optical clocks and relativity. *Science* 329, 1630–1633 (2010).
- 415 3. Herrmann, S. *et al.* Test of the Gravitational Redshift with Galileo Satellites in an Eccentric Orbit.
 416 *Phys. Rev. Lett.* **121**, 231102 (2018).
- 4. Delva, P. *et al.* Gravitational Redshift Test Using Eccentric Galileo Satellites. *Phys. Rev. Lett.*418 121, 231101 (2018).
- 419 5. Campbell, S. L. *et al.* A Fermi-degenerate three-dimensional optical lattice clock. *Science* 358, 90–94 (2017).
- 421 6. Oelker, E. *et al.* Demonstration of 4.8×10^{-17} stability at 1 s for two independent optical clocks. 422 *Nat. Photonics* **13**, 714–719 (2019).
- 423 7. Nicholson, T. *et al.* Systematic evaluation of an atomic clock at 2×10^{-18} total uncertainty. *Nat.* 424 *Commun.* **6**, (2015).
- 425 8. McGrew, W. F. *et al.* Atomic clock performance enabling geodesy below the centimetre level.
 426 *Nature* 564, 87–90 (2018).
- 427 9. Brewer, S. M. *et al.* ²⁷Al⁺ Quantum-Logic Clock with a Systematic Uncertainty below 10^{-18} . *Phys.* 428 *Rev. Lett.* **123**, 33201 (2019).
- 429 10. Bothwell, T. *et al.* JILA SrI optical lattice clock with uncertainty of 2.0×10^{-18} . *Metrologia* 56, (2019).
- 431 11. Marti, G. E. *et al.* Imaging Optical Frequencies with 100 μHz Precision and 1.1 μm Resolution.
 432 *Phys. Rev. Lett.* **120**, (2018).
- 433 12. Pedrozo-peñafiel, E. *et al.* Entanglement on an optical atomic-clock transition. *Nature* 588, (2020).
- 435 13. Kaubruegger, R. *et al.* Variational Spin-Squeezing Algorithms on Programmable Quantum
 436 Sensors. *Phys. Rev. Lett.* **123**, 260505 (2019).
- 437 14. Kómár, P. *et al.* Quantum Network of Atom Clocks: A Possible Implementation with Neutral
 438 Atoms. *Phys. Rev. Lett.* **117**, 060506 (2016).
- 439 15. Young, A. W. *et al.* Half-minute-scale atomic coherence and high relative stability in a tweezer clock. *Nature* 588, 408–413 (2020).
- 441 16. Safronova, M. S. *et al.* Search for new physics with atoms and molecules. *Rev. Mod. Phys.* 90, 25008 (2018).
- 443 17. Sanner, C. *et al.* Optical clock comparison for Lorentz symmetry testing. *Nature* 567, 204–208 (2019).
- 18. Kennedy, C. J. et al. Precision Metrology Meets Cosmology: Improved Constraints on Ultralight

446		Dark Matter from Atom-Cavity Frequency Comparisons. Phys. Rev. Lett. 125, 1-10 (2020).	
447 448	19.	Boulder Atomic Clock Optical Network. Frequency ratio measurements at 18-digit accuracy using an optical clock network. 591 , (2021).	
449 450	20.	Kolkowitz, S. <i>et al.</i> Gravitational wave detection with optical lattice atomic clocks. <i>Phys. Rev. D</i> 94 , 124043 (2016).	
451	21.	Hafele, J. C. & Keating, R. E. Around-the-World Atomic Clocks: Science 177, 166 (1972).	
452 453	22.	Takamoto, M. <i>et al.</i> Test of general relativity by a pair of transportable optical lattice clocks. <i>Nat. Photonics</i> 14 , 411–415 (2020).	
454 455	23.	Laurent, P., Massonnet, D., Cacciapuoti, L. & Salomon, C. The ACES/PHARAO space mission. <i>Comptes Rendus Phys.</i> 16 , 540–552 (2015).	
456 457	24.	Tino, G. M. <i>et al.</i> SAGE : A proposal for a space atomic gravity explorer. <i>Eur. Phys. J. D</i> 73 , 228 (2019).	
458 459	25.	Grotti, J. <i>et al.</i> Geodesy and metrology with a transportable optical clock. <i>Nat. Phys.</i> 14 , 437–441 (2018).	
460 461	26.	Flechtner, Frank, Nico Sneeuw, and Wolf-Dieter Schuh. Observation of the system earth from space: CHAMP, GRACE, GOCE and future missions. <i>Springer</i> (2014).	
462 463	27.	Kolkowitz, S. <i>et al.</i> Spin-orbit-coupled fermions in an optical lattice clock. <i>Nature</i> 542 , 66–70 (2017).	
464 465	28.	Bromley, S. L. <i>et al.</i> Dynamics of interacting fermions under spin-orbit coupling in an optical lattice clock. <i>Nat. Phys.</i> 14 , 399–404 (2018).	
466 467 468	29.	Wilkinson, S. R., Bharucha, C. F., Madison, K. W., Niu, Q. & Raizen, M. G. Observation of atomic wannier-stark ladders in an accelerating optical potential. <i>Phys. Rev. Lett.</i> 76 , 4512–4515 (1996).	
469 470	30.	Lemonde, P. & Wolf, P. Optical lattice clock with atoms confined in a shallow trap. <i>Phys. Rev. A</i> - <i>At. Mol. Opt. Phys.</i> 72 , 1–8 (2005).	
471	31.	Aeppli, A. et al., In Preparation (2021).	
472 473 474	32.	Muniz, J. A., Young, D. J., Cline, J. R. K. & Thompson, J. K. Cavity-QED measurements of the ⁸⁷ Sr millihertz optical clock transition and determination of its natural linewidth . <i>Phys. Rev. Res.</i> 3 , 023152 (2021).	
475 476	33.	Ludlow, A. D., Boyd, M. M., Ye, J., Peik, E. & Schmidt, P. O. Optical atomic clocks. <i>Rev. Mod. Phys.</i> 87, 637–701 (2015).	
477 478 479	34.	Matei, D. G. et al. 1.5 µm Lasers with Sub-10 mHz Linewidth. Phys. Rev. Lett. 118, 263202 (2017).	
480	References - Methods		
481	35	Lemonde P Brusch A Targat R Le Baillard X & Fouche M Hyperpolarizability Effects in	

481 35. Lemonde, P., Brusch, A., Targat, R. Le, Baillard, X. & Fouche, M. Hyperpolarizability Effect
482 a Sr Optical Lattice Clock. *Phys. Rev. Lett.* 96, 103003 (2006).

- 483 36. Lodewyck, J., Zawada, M., Lorini, L., Gurov, M. & Lemonde, P. Observation and Cancellation of
 484 a Perturbing dc Stark Shift in Strontium Optical Lattice Clocks. *IEEE Transactions on*485 *Ultrasonics, Ferroelectrics, and Frequency Control.* 59, 411–415 (2012).
- 486 37. Leibfried, D., Blatt, R., Monroe, C. & Wineland, D. Quantum dynamics of single trapped ions.
 487 *Rev. Mod. Phys.* 75, 281–324 (2003).
- 488 38. Marti, G. E. *et al.* Imaging Optical Frequencies with 100 μhz Precision and 1.1 μ m Resolution.
 489 *Phys. Rev. Lett.* **120**, 103201 (2018).
- 490 39. Boyd, M. M. et al. Nuclear spin effects in optical lattice clocks. Phys. Rev. A 76, 022510 (2007).
- 40. Martin, M. J. *et al.* A quantum many-body spin system in an optical lattice clock. *Science* 341, 632–636 (2013).
- 41. Ushijima, I. *et al.* Operational Magic Intensity for Sr Optical Lattice Clocks. *Phys. Rev. Lett.* 121, 263202 (2018).
- 495 42. van Westrum, D. NOAA Technical Memorandum NOS NGS-77. (2019).
- 43. Xin Zheng Jonathan Dolde, V. L. B. M. H. L. & Kolkowitz, S. High precision differential clock
 497 comparisons with a multiplexed optical lattice clock (submitted 2021).
- 498









