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# Resolving the gravitational redshift within a millimeter atomic sample

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**Einstein's theory of general relativity states that clocks at different gravitational potentials tick at different rates – an effect known as the gravitational redshift<sup>1</sup>. As fundamental probes of space and time, atomic clocks have long served to test this prediction at distance scales from 30 centimeters to thousands of kilometers<sup>2-4</sup>. Ultimately, clocks will study the union of general relativity and quantum mechanics once they become sensitive to the finite wavefunction of quantum objects oscillating in curved spacetime. Towards this regime, we measure a linear frequency gradient consistent with the gravitational redshift within a single millimeter scale sample of ultracold strontium. Our result is enabled by improving the fractional frequency measurement uncertainty by more than a factor of 10, now reaching  $7.6 \times 10^{-21}$ . This heralds a new regime of clock operation necessitating intra-sample corrections for gravitational perturbations.**

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-19<sup>th</sup> digit in one

27 hour<sup>5,6</sup>, and three atomic species achieved systematic uncertainties corresponding to an error  
28 equivalent to less than 1 s over the lifetime of the Universe<sup>7-10</sup>. Central to this success in  
29 neutral atom clocks is the ability to maintain extended quantum coherence times while using  
30 large ensembles of atoms<sup>5,6,11</sup>. The pace of progress has yet to slow. Continued improvement  
31 in measurement precision and accuracy arising from the confluence of metrology and quantum  
32 information science<sup>12-15</sup> promises discoveries in fundamental physics<sup>16-20</sup>.

33 Clocks fundamentally connect space and time, providing exquisite tests of the theory of  
34 general relativity. Hafele and Keating took cesium-beam atomic clocks aboard commercial  
35 airliners in 1971, observing differences between flight-based and ground-based clocks  
36 consistent with special and general relativity<sup>21</sup>. More recently, RIKEN researchers compared  
37 two strontium optical lattice clocks (OLCs) separated by 450 m in the Tokyo Skytree, resulting  
38 in the most precise terrestrial redshift measurement to date<sup>22</sup>. Proposed satellite-based  
39 measurements<sup>23,24</sup> will provide orders of magnitude improvement to current bounds on  
40 gravitational redshifts<sup>3,4</sup>. Concurrently, clocks are anticipated to begin playing important roles  
41 for relativistic geodesy<sup>25</sup>. In 2010 Chou *et al.*<sup>2</sup> demonstrated the precision of their Al<sup>+</sup> clocks by  
42 measuring the gravitational redshift resulting from lifting one clock vertically by 30 cm in 40  
43 hours. With a decade of advancements, today's leading clocks are poised to enable local  
44 geodetic surveys of elevation at the sub-centimeter level on Earth, complimenting spatial  
45 averaging techniques<sup>26</sup>.

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

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

61 Central to our experiment is an in-vacuum optical cavity (Fig. 1a and Methods) for  
62 power enhancement of the optical lattice. The cavity (finesse 1300) ensures wavefront  
63 homogeneity of our 1D lattice while the large beam waist (260  $\mu\text{m}$ ) reduces the atomic density  
64 by an order of magnitude compared to our previous system<sup>10</sup>. We begin each experiment by  
65 trapping fermionic  $^{87}\text{Sr}$  atoms into the 1D lattice at a trap depth of 300 lattice photon recoil  
66 energies ( $E_{\text{rec}}$ ), loading a millimeter scale atomic sample (Fig. 1a). Atoms are simultaneously  
67 cooled and polarized into a single nuclear spin before the lattice is adiabatically ramped to an  
68 operational depth of  $12 E_{\text{rec}}$ . Clock interrogation proceeds by probing the ultranarrow  $^1\text{S}_0(g) \rightarrow$   
69  $^3\text{P}_0(e)$  transition with the resulting excitation fraction measured by fluorescence spectroscopy.  
70 Scattered photons are collected on a camera, enabling *in-situ* measurement with 6  $\mu\text{m}$   
71 resolution, corresponding to  $\sim 15$  lattice sites.

72 Quantum state control has been vital to recent advances in atom-atom and atom-light  
73 coherence times in 3D OLCs and tweezer clocks<sup>5,11,15</sup>. Improved quantum state control is  
74 demonstrated through precision spectroscopy of the Wannier-Stark states of the OLC<sup>29,30</sup>. The  
75 1D lattice oriented along gravity has the degeneracy of neighboring lattice sites lifted by the  
76 gravitational potential energy. In the limit of shallow lattice depths, this creates a set of  
77 delocalized states. By ramping the lattice depth to  $6 E_{\text{rec}}$ , much lower than in traditional 1D  
78 lattice operations<sup>7,8,10</sup>, clock spectroscopy probes this delocalization (Fig. 1d). The ability to  
79 engineer the extent of atomic wavefunctions through the adjustment of trap depth creates an  
80 opportunity to control the balance of on-site *p*-wave versus neighboring-site *s*-wave atomic  
81 interactions. We utilize this tunability by operating at a ‘magic’ trap depth<sup>31</sup>, where the  
82 frequency shifts arising from on-site and off-site atomic interactions cancel, enabling a  
83 reduction of the collisional frequency shifts by more than three orders of magnitude compared  
84 our previous work<sup>10</sup>.

85 Extended atomic coherence times are critical for both accuracy and precision. An  
86 aspirational milestone for clock measurement precision is the ability to coherently interrogate  
87 atomic samples up to the excited state’s natural lifetime. To evaluate the limits of our clock’s  
88 atomic coherence, we perform Ramsey spectroscopy to measure the decay of fringe contrast  
89 as a function of the free-evolution time. By comparing two uncorrelated regions within our  
90 atomic sample, we determine the contrast and relative phase difference between the two sub-  
91 ensembles (Fig. 2). The contrast decays exponentially with a time constant of 37 s (quality  
92 factor of  $3.6 \times 10^{16}$ ), corresponding to an additional decoherence time of 53 s relative to the  $^3\text{P}_0$   
93 natural lifetime (118 s)<sup>32</sup>. This represents the longest optical atomic coherence time measured  
94 in any spectroscopy system to date.

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

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 ( $\delta f$ ) as given by

116 
$$\frac{\delta f}{f} = \frac{ah}{c^2} \quad (\text{Eq. 1}),$$

117 with  $f$  the clock frequency,  $c$  the speed of light, and  $a$  the gravitational acceleration. The  
118 gravitational redshift at Earth's surface corresponds to a fractional frequency gradient of  $-1.09 \times$   
119  $10^{-19}/\text{mm}$  in the coordinate system of Fig. 1a. Measurement of a vertical gradient across the  
120 atomic sample consistent with the gravitational redshift provides an exquisite verification of an  
121 individual atomic clock's frequency control.

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

130 The ability to resolve the gravitational redshift within our system suggests a level of  
131 frequency control beyond previous clock demonstrations, vital for the continued advancement  
132 of clock accuracy and precision. Previous fractional frequency comparisons<sup>15</sup> have reached  
133 uncertainties as low as  $4.2 \times 10^{-19}$ . Similarly, we perform a synchronous comparison between  
134 two uncorrelated regions of our atomic cloud (Fig. 4a). By binning  $\sim 100$  pixels per region, we  
135 substantially reduce instability caused by quantum projection noise<sup>33</sup>. Analyzing the frequency  
136 difference between regions from 92 hours of data, we find a fractional frequency instability of  
137  $4.4 \times 10^{-18}/\sqrt{\tau}$  ( $\tau$  is the averaging time in seconds), resulting in a fractional frequency uncertainty  
138 of  $7.6 \times 10^{-21}$  for full measurement time, nearly two orders of magnitude lower than the previous  
139 record. From this measurement we infer a single region instability of  $3.1 \times 10^{-18}/\sqrt{\tau}$ . Dividing the

140 fractional frequency difference by the spatial separation between each region's center of mass  
141 gives a frequency gradient of  $-1.30(18)\times 10^{-19}/\text{mm}$ . Correcting for additional systematics as  
142 before results in a gradient of  $-1.28(27)\times 10^{-19}/\text{mm}$ , again fully consistent with the predicted  
143 redshift.

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



153 **Fig. 1: Experimental system and quantum state control.** **a**, A millimeter length sample of  
154  $\sim 100,000$   $^{87}\text{Sr}$  atoms are trapped in a 1D optical lattice formed within an in-vacuum cavity. The  
155 longitudinal axis of the cavity,  $z$ , is oriented along gravity. We probe atoms along the  $^1\text{S}_0 \rightarrow ^3\text{P}_0$   
156 transition using a clock laser locked to an ultrastable crystalline silicon cavity<sup>6,34</sup>. **b**, Rabi  
157 spectroscopy with a 3.1 s pulse time. Open purple circles indicate data with a corresponding  
158 Rabi fit in green. **c**, Neighboring lattice sites are detuned by gravity, creating a Wannier-Stark  
159 ladder. Clock spectroscopy probes the overlap of Wannier-Stark states between lattice sites  
160 that are  $m$  sites away with Rabi frequency  $\Omega_m$ . **d**, Rabi spectroscopy probes Wannier-Stark  
161 state transitions, revealing wavefunction delocalization of up to 5 lattice sites. The number of  
162 lattice sites is indicated above each transition, with blue(red) denoting Wannier-Stark  
163 transitions to higher(lower) lattice sites.

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166 **Fig. 2: Atomic coherence.** We use Ramsey spectroscopy with a randomly sampled phase for  
167 the second pulse to determine the coherence time of our system<sup>11</sup>. **a**, We measure the  
168 excitation fraction across the cloud, shown in purple for a single measurement, and calculate  
169 the average excitation fractions in regions  $p_1$  and  $p_2$ , separated by 2 pixels. **b**, Parametric plots  
170 of the excitation fraction of  $p_1$  versus  $p_2$  in purple for 6 s, 30 s and 50 s dark time demonstrate  
171 a phase shift between the two regions and contrast decay. Using a maximum likelihood  
172 estimator, we extract the phase and contrast for each dark time with the fit, shown in green. **c**,  
173 Contrast decay as a function of time in green is fit with an exponential decay in gold, giving an  
174 atomic coherence decay time of 36.5(0.7) s and a corresponding quality factor of  $3.6 \times 10^{16}$ .  
175 After accounting for the finite radiative decay contribution, we infer an additional decoherence  
176 time constant of 52.8(1.5) s.

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

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

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## Methods

### In-Vacuum Cavity

Central to our system is an in-vacuum lattice buildup cavity oriented along gravity (Fig. 1a). Two mirrors with radius of curvature of 1 m are separated by  $\sim 15$  cm, achieving a mode waist of  $260 \mu\text{m}$ . Our over-coupled cavity has a finesse at the lattice wavelength (813 nm) of  $\sim 1300$  and a power buildup factor of  $\sim 700$  (ratio of circulating to input intensity). This enables lattice depths in excess of  $500 E_{\text{rec}}$  (lattice photon recoil energy) using a diode-based laser system. The dimensional stability of the cavity combined with the simplified diode laser system enables robust operation compared with our previous Ti:Sapphire retro-reflected design<sup>10</sup>. The cavity mirrors are anti-reflection coated at the clock wavelength of 698 nm.

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

### Atomic sample preparation

<sup>87</sup>Sr atoms are cooled and loaded into a  $300 E_{\text{rec}}$  optical lattice using standard two stage magneto-optical trapping techniques<sup>10</sup>. 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  $12 E_{\text{rec}}$ , where a series of pulses addressing the clock transition prepares atoms into

232  $|g, m_F = \pm \frac{5}{2}\rangle$ . Clock spectroscopy is performed by interrogating the  $|g, m_F = \pm \frac{5}{2}\rangle$  to  $|e, m_F =$   
233  $\pm \frac{3}{2}\rangle$  transition, the most magnetically insensitive  $^{87}\text{Sr}$  clock transition<sup>6</sup>.

## 234 **Imaging**

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

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

## 244 **Analysis**

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

253 clock lock, a pixel specific center frequency  $f_i$  and frequency splitting between opposite  $m_F$   
254 states  $\Delta_i$  are calculated, creating an *in-situ* frequency map of the 1D atomic sample. This  
255 allows rejection of vector shifts on a pixel-by-pixel basis and probes the magnetic field induced  
256 splitting of  $m_F$  transitions. The atom weighted mean frequency is subtracted from every lock  
257 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 ( $\sigma$ ). All analysis is performed within the central  
260 region of  $\pm 1.5\sigma$  which demonstrates the lowest frequency instability. Identifying a center pixel  
261 for data processing ensures rejection of any day-to-day drift in the position of the cloud due to  
262 varying magnetic fields modifying MOT operation on the narrow line transition. The density  
263 shift coefficient (see Density Shift section) is derived from the average center frequency per  
264 pixel. Using this coefficient, we correct  $f_i$  and  $\Delta_i$  for the density shift. 2<sup>nd</sup> order Zeeman  
265 corrections using these updated frequencies are then applied.

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

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

## 276 **Atomic Coherence**

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

281 Two regions,  $p_1$  and  $p_2$ , are identified using the same technique as in the synchronous  
282 instability measurement. For each experimental sequence, we find the average excitation  
283 fraction in  $p_1$  and  $p_2$ . A mean frequency shift across the sample primarily due to a magnetic  
284 field gradient creates a differential phase as a function of time between  $p_1$  and  $p_2$ . We create a  
285 parametric plot of the average excitation in  $p_1$  and  $p_2$  for each dark time and use a maximum  
286 likelihood estimator to fit an ellipse to each dataset, calculating phase and contrast<sup>15,38</sup>. To  
287 estimate uncertainty in the contrast for each dark time a bootstrapping technique is used<sup>11</sup>.  
288 Fitting the contrast as a function of dark time with a single exponential returns an effective  
289 atomic coherence time.

## 290 **Systematics**

### 291 **Imaging**

292 We calibrate our pixel size using standard time of flight methods: we observe an atomic  
293 sample in freefall for varying times to determine an effective pixel size along the direction of  
294 gravity. Immediately after our 10-day data campaign we measured our effective pixel size to be  
295  $6.04 \mu\text{m}$ . Due to thermal drift of our system, the pixel size can vary by up to  $0.5 \mu\text{m} / \text{pixel}$  over  
296 months which we take as the calibration uncertainty.



297 Spatial correlations may limit imaging resolution. We measure these correlations by  
298 placing atoms into a superposition of clock states. Any measured spatial correlation is due to  
299 the imaging procedure. In our system we find no correlations between neighboring pixels<sup>11</sup>.  
300 The optical resolution of our imaging lens is specified at 2  $\mu\text{m}$ .

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

### 304 Zeeman Shifts

305 First order Zeeman shifts are rejected by probing opposite nuclear spin states<sup>6</sup>. The 2<sup>nd</sup>  
306 order Zeeman shift is given by

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

307 where  $\Delta\nu_{B,1}$  is the splitting between opposite spin states and  $\xi$  the corresponding 2<sup>nd</sup> order  
308 Zeeman shift coefficient. For stretched spin state operation ( $m_F = \pm\frac{9}{2}$ ),  $\xi = -2.456(3) \times 10^{-7} \text{ Hz}^{-1}$ .  
309 Using known atomic coefficients<sup>39</sup> we find the 2<sup>nd</sup> order Zeeman coefficient for the  $|g, m_F =$   
310  $\pm\frac{5}{2}\rangle$  to  $|e, m_F = \pm\frac{3}{2}\rangle$  transition to be  $\xi_{op} = -1.23(8) \times 10^{-4} \text{ Hz}$ , with the uncertainty arising from  
311 limited knowledge of atomic coefficients.

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

## 317 **DC Stark**

318 Electric fields perturb the clock frequency via the DC Stark effect. We evaluate  
319 gradients arising from this shift by using in-vacuum quadrant electrodes to apply bias electric  
320 fields in all three dimensions. We find a DC Stark gradient of  $3(2)\times 10^{-21}/\text{mm}$ .

## 321 **Black Body Radiation Shift**

322 The dominant frequency perturbation to room temperature neutral atom clocks is black  
323 body radiation (BBR). Similar to our previous work<sup>10</sup>, we homogenize this shift by carefully  
324 controlling the thermal surroundings of our vacuum chamber. Attached to the vacuum chamber  
325 are additional temperature control loops, with each vacuum viewport having a dedicated  
326 temperature control system. This ensures our dominant BBR contribution – high emissivity  
327 glass viewports – are all the same temperature to within 100 mK.

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

## 334 **Density Shift**

335 Atomic interactions during Rabi spectroscopy lead to clock frequency shifts as a  
336 function of atomic density<sup>40</sup>. For each gradient measurement, we evaluate the density shift  
337 coefficient  $\chi_{dens}$  by fitting the average frequency  $f$  per pixel versus average camera counts per  
338 pixel  $N$  to an equation of the form

339 
$$f(N) = \chi_{dens}N + B.$$

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

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

347 **Lattice Light Shifts**

348 Lattice light shifts arise from differential AC Stark shifts between the ground and excited  
 349 clock states. An approximate microscopic model of the lattice light shift ( $\nu_{LS}$ ) in our system is  
 350 given by<sup>41</sup>

351 
$$h\nu_{LS}(u, \delta_L) \approx \left( \frac{\delta\Delta\alpha^{E1}}{\delta\nu} \delta_L - \Delta\alpha_{QM} \right) \frac{u^2}{2} - \left[ \frac{\delta\alpha^{E1}}{\delta\nu} \delta_L \right] u,$$

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

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

$$360 \quad \frac{\delta h\nu_{LS}(u, \delta_L)}{\delta z} \approx \left[ \frac{\left( \frac{\delta \alpha^{E1}}{\delta \nu} \delta_L - \alpha_{QM} \right)}{4u^{1/2}} - \frac{\delta \alpha^{E1}}{\delta \nu} \delta_L \right] \frac{\delta u}{\delta z},$$

361 where  $z$  is the coordinate corresponding to the axis of the cavity along gravity. To evaluate our  
 362 differential lattice light shift at our operational depth we need  $\delta_L$  and  $\frac{\delta u}{\delta z}$ . We modulate our  
 363 lattice between two trap depths ( $u_1=14 E_{\text{rec}}$ ,  $u_2=56 E_{\text{rec}}$ ) and find our detuning from scalar  
 364 magic frequency to be  $\delta_L=7.4(0.6)$  MHz. To evaluate  $\frac{\delta u}{\delta z}$  at our operational depth ( $u_{op}$ ) we  
 365 measure the linear gradient across the atomic cloud at  $\delta_L + 250$  MHz and  $\delta_L - 250$  MHz, the  
 366 difference given by

$$367 \quad \frac{\delta h\nu_{LS}(u_{op}, \delta_L + 250 \text{ MHz})}{\delta z} - \frac{\delta h\nu_{LS}(u_{op}, \delta_L - 250 \text{ MHz})}{\delta z} \approx \left[ \frac{1}{4u_{op}^2} - 1 \right] \frac{\delta \alpha^{E1}}{\delta \nu} \delta_{500} \frac{\delta u_{op}}{\delta z},$$

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

### 371 Other Systematics

372 For a 3.1 s  $\pi$ -pulse the probe AC Stark shift<sup>7</sup> is  $-3(2) \times 10^{-21}$ . A frequency scan of the  $|g, m_F =$   
 373  $-\frac{5}{2}\rangle$  to  $|e, m_F = -\frac{3}{2}\rangle$  transition limits the variation of excitation fraction across the atomic  
 374 sample to 1% or below, bounding any possible probe AC Stark gradient across the sample to  
 375  $< 1 \times 10^{-22}$ .

### 376 Known Redshift

377 The gravitational acceleration (rounding to 4 digits) within our lab was evaluated by a  
 378 USGS survey<sup>42</sup> to be  $a=-9.796 \text{ m/s}^2$ .

379 **Systematic Budget**

Systematic	Slope ( $10^{-20}/\text{mm}$ )	Uncertainty ( $10^{-20}/\text{mm}$ )
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 <sup>nd</sup> Order Zeeman	-	.4
Other	0	<.1
Corrected Gradient	-9.8	2.3
Known Redshift	-10.9	<.1

380

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

385

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395 **Authors' note:** While performing the work described here, we became aware of  
396 complementary work where high measurement precision was achieved for simultaneous  
397 differential clock comparisons between multiple atomic ensembles in vertical 1D  
398 lattices separated by centimeter scale distances using a hertz-linewidth clock laser<sup>43</sup>.

#### 399 **Author Contributions**

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

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#### 403 **Competing interests**

404 The authors declare no competing interests.

#### 405 **Data and Code Availability**

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

408

409 **References**

410

- 411 1. Einstein, A. Grundgedanken der allgemeinen Relativitätstheorie und Anwendung dieser Theorie  
412 in der Astronomie. *Preuss. Akad. der Wissenschaften, Sitzungsberichte* **315**, (1915).
- 413 2. Chou, C. W., Hume, D. B., Rosenband, T. & Wineland, D. J. Optical clocks and relativity.  
414 *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).
- 417 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**,  
420 90–94 (2017).
- 421 6. Oelker, E. *et al.* Demonstration of  $4.8 \times 10^{-17}$  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  $2 \times 10^{-18}$  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.*  $^{27}\text{Al}^+$  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  $2.0 \times 10^{-18}$ . *Metrologia* **56**,  
430 (2019).
- 431 11. Marti, G. E. *et al.* Imaging Optical Frequencies with 100  $\mu\text{Hz}$  Precision and 1.1  $\mu\text{m}$  Resolution.  
432 *Phys. Rev. Lett.* **120**, (2018).
- 433 12. Pedrozo-peñañiel, E. *et al.* Entanglement on an optical atomic-clock transition. *Nature* **588**,  
434 (2020).
- 435 13. Kaubuegger, 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  
440 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**,  
442 25008 (2018).
- 443 17. Sanner, C. *et al.* Optical clock comparison for Lorentz symmetry testing. *Nature* **567**, 204–208  
444 (2019).
- 445 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 19. Boulder Atomic Clock Optical Network. Frequency ratio measurements at 18-digit accuracy  
448 using an optical clock network. **591**, (2021).
- 449 20. Kolkowitz, S. *et al.* Gravitational wave detection with optical lattice atomic clocks. *Phys. Rev. D*  
450 **94**, 124043 (2016).
- 451 21. Hafele, J. C. & Keating, R. E. Around-the-World Atomic Clocks: *Science* **177**, 166 (1972).
- 452 22. Takamoto, M. *et al.* Test of general relativity by a pair of transportable optical lattice clocks. *Nat.*  
453 *Photonics* **14**, 411–415 (2020).
- 454 23. Laurent, P., Massonnet, D., Cacciapuoti, L. & Salomon, C. The ACES/PHARAO space mission.  
455 *Comptes Rendus Phys.* **16**, 540–552 (2015).
- 456 24. Tino, G. M. *et al.* SAGE : A proposal for a space atomic gravity explorer. *Eur. Phys. J. D* **73**, 228  
457 (2019).
- 458 25. Grotti, J. *et al.* Geodesy and metrology with a transportable optical clock. *Nat. Phys.* **14**, 437–441  
459 (2018).
- 460 26. Flechtner, Frank, Nico Sneeuw, and Wolf-Dieter Schuh. Observation of the system earth from  
461 space: CHAMP, GRACE, GOCE and future missions. *Springer* (2014).
- 462 27. Kolkowitz, S. *et al.* Spin-orbit-coupled fermions in an optical lattice clock. *Nature* **542**, 66–70  
463 (2017).
- 464 28. Bromley, S. L. *et al.* Dynamics of interacting fermions under spin-orbit coupling in an optical  
465 lattice clock. *Nat. Phys.* **14**, 399–404 (2018).
- 466 29. Wilkinson, S. R., Bharucha, C. F., Madison, K. W., Niu, Q. & Raizen, M. G. Observation of  
467 atomic wannier-stark ladders in an accelerating optical potential. *Phys. Rev. Lett.* **76**, 4512–4515  
468 (1996).
- 469 30. Lemonde, P. & Wolf, P. Optical lattice clock with atoms confined in a shallow trap. *Phys. Rev. A*  
470 - *At. Mol. Opt. Phys.* **72**, 1–8 (2005).
- 471 31. Aeppli, A. *et al.*, In Preparation (2021).
- 472 32. Muniz, J. A., Young, D. J., Cline, J. R. K. & Thompson, J. K. Cavity-QED measurements of the  
473 <sup>87</sup>Sr millihertz optical clock transition and determination of its natural linewidth . *Phys. Rev. Res.*  
474 **3**, 023152 (2021).
- 475 33. Ludlow, A. D., Boyd, M. M., Ye, J., Peik, E. & Schmidt, P. O. Optical atomic clocks. *Rev. Mod.*  
476 *Phys.* **87**, 637–701 (2015).
- 477 34. Matei, D. G. *et al.* 1.5  $\mu$ m Lasers with Sub-10 mHz Linewidth. *Phys. Rev. Lett.* **118**, 263202  
478 (2017).  
479

## 480 References - Methods

- 481 35. Lemonde, P., Bruschi, A., Targat, R. Le, Baillard, X. & Fouche, M. Hyperpolarizability Effects in  
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  $\mu\text{Hz}$  Precision and 1.1  $\mu\text{m}$  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).
- 491 40. Martin, M. J. *et al.* A quantum many-body spin system in an optical lattice clock. *Science* **341**,  
492 632–636 (2013).
- 493 41. Ushijima, I. *et al.* Operational Magic Intensity for Sr Optical Lattice Clocks. *Phys. Rev. Lett.* **121**,  
494 263202 (2018).
- 495 42. van Westrum, D. NOAA Technical Memorandum NOS NGS-77. (2019).
- 496 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).
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