

Blackbody radiation shift of the  $^{27}\text{Al}^+ \ ^1\text{S}_0 \rightarrow \ ^3\text{P}_0$  transition\*T. Rosenband,<sup>†</sup> W. M. Itano, P. O. Schmidt,<sup>‡</sup> D. B. Hume,J. C. J. Koelemeij,<sup>§</sup> J. C. Bergquist, and D. J. Wineland

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The differential polarizability, due to near-infrared light at 1126 nm, of the  $^{27}\text{Al}^+ \ ^1\text{S}_0 \rightarrow \ ^3\text{P}_0$  transition is measured to be  $\Delta\alpha = 4\pi\epsilon_0 \times (1.6 \pm 0.5) \times 10^{-31} \text{ m}^2$ , where  $\Delta\alpha = \alpha_P - \alpha_S$  is the difference between the excited and ground state polarizabilities. This measurement is combined with experimental oscillator strengths to extrapolate the differential static polarizability of the clock transition as  $\Delta\alpha(0) = 4\pi\epsilon_0 \times (1.5 \pm 0.5) \times 10^{-31} \text{ m}^2$ . The resulting room temperature blackbody shift of  $\Delta\nu/\nu = -8(3) \times 10^{-18}$  is the lowest known shift of all atomic transitions under consideration for optical frequency standards. A method is presented to estimate the differential static polarizability of an optical transition, from a differential light shift measurement.

The blackbody radiation shift [1] is a significant shift in all room temperature atomic frequency standards, as can be seen in Table I. It ranges from  $|\Delta\nu/\nu| \approx 2 \times 10^{-14}$  for  $^{133}\text{Cs}$  to  $|\Delta\nu/\nu| \approx 8 \times 10^{-18}$  for  $^{27}\text{Al}^+$ , as reported here. In order to reach a systematic uncertainty of  $|\Delta\nu/\nu| < 10^{-18}$ , the transitions with a large room temperature blackbody shift may require a cryogenic operating environment, while  $^{27}\text{Al}^+$  merely requires knowledge of the room temperature background with 5 K uncertainty.

We begin with a brief explanation of the blackbody shift, followed by an estimate of the shift in  $^{27}\text{Al}^+$  based only on published oscillator strengths. The uncertainty in this estimate motivated us to measure the differential polarizability of the clock transition due to

near-infrared light. This measurement allows a determination of the blackbody shift with reduced uncertainty.

## A. Blackbody shift

The blackbody shift results from off-resonant coupling of thermal blackbody radiation to the two states comprising the clock transition. The scalar polarizability  $\alpha_a$  of an atomic state  $a$  driven by an electric field at frequency  $\omega$  is

$$\alpha_a(\omega) = \frac{e^2}{m_e} \sum_i \frac{f_i}{\omega_i^2 - \omega^2}, \quad (1)$$

TABLE I: Room temperature blackbody shifts and uncertainties of various species in use, or under consideration, as atomic frequency standards. Where no uncertainty is given, it is unknown. The  $^{199}\text{Hg}^+$  optical transition is not listed, because this standard operates at 4.2 K, where the blackbody shift is reduced by  $10^7$  from the room temperature value.

species	transition	$ \Delta\nu/\nu  \times 10^{-18}$	reference
Al <sup>+</sup>	$^1\text{S}_0 \rightarrow ^3\text{P}_0$	8(3)	<i>this work</i>
In <sup>+</sup>	$^1\text{S}_0 \rightarrow ^3\text{P}_0$	< 70	[2]
Yb <sup>+</sup>	$^2\text{S}_{1/2} \rightarrow ^2\text{D}_{3/2}$	580(30)	[3]
Sr <sup>+</sup>	$^2\text{S}_{1/2} \rightarrow ^2\text{D}_{5/2}$	670(250)	[4]
Ca	$^1\text{S}_0 \rightarrow ^3\text{P}_1$	2210(50)	[5]
Yb	$^1\text{S}_0 \rightarrow ^3\text{P}_0$	2400(250)	[6]
Sr	$^1\text{S}_0 \rightarrow ^3\text{P}_0$	5100(120)	[7]
Cs	F=4 → F=3	21210(260)	[8]

with summation over all transitions connecting to state  $a$  with resonant frequency  $\omega_i$ , and oscillator strength  $f_i$ . For monochromatic radiation  $E_0 \cos \omega t$ , this polarizability results in a dynamic Stark shift of  $\Delta E_a = -\frac{1}{4} E_0^2 \alpha_a(\omega)$ . The clock transition is subject to a blackbody radiation shift of

$$\Delta\nu = \frac{-1}{4\epsilon_0\pi^3 c^3} \int_0^\infty \Delta\alpha(\omega) \frac{\omega^3}{e^{h\omega/k_B T} - 1} d\omega, \quad (2)$$

where we integrate over the power spectral density of the blackbody electric field, and  $\Delta\alpha(\omega) = \alpha_P(\omega) - \alpha_S(\omega)$  is the difference between excited and ground state polarizabilities. Here, we first estimate the differential static polarizability  $\Delta\alpha(0)$ . This result is used to estimate the differential polarizability at blackbody frequencies  $\Delta\alpha(\omega)$  for  $\omega \approx 2\pi c/(10 \mu\text{m})$ .

B. The case of  $^{27}\text{Al}^+$ 

The transitions from the  $^1\text{S}_0$  and  $^3\text{P}_0$  states that have been included in our estimate are listed in Table II. Oscillator strengths are taken from the *NIST Atomic Spectra Database* [9] where available, and from the *Opacity Project* [10] otherwise. From this we calculate for the  $^3\text{P}_0$  state  $\alpha_P(0) = 4\pi\epsilon_0 \times 3.65(73) \times 10^{-30} \text{ m}^2$ . For the  $^1\text{S}_0$  state  $\alpha_S(0) = 4\pi\epsilon_0 \times 3.68(78) \times$

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$10^{-30}$  m<sup>3</sup>. Thus,  $\Delta\alpha(0) = 4\pi\epsilon_0 \times (-0.03 \pm 1.0) \times 10^{-30}$  m<sup>3</sup>. The room temperature blackbody spectrum ( $E_{rms} = 830$  V/m) is centered at 10  $\mu$ m wavelength. This corresponds to a frequency  $\omega$  in Eq. (1), which is 50 times lower than the lowest transition frequency  $\omega_i$ . We may use the static polarizability without loss of accuracy, and find  $\Delta\nu = -\frac{1}{2h}\Delta\alpha(0)E_{rms}^2 = (0.00 \pm 0.06)$  Hz, or fractionally  $\Delta\nu/\nu = (0 \pm 6) \times 10^{-17}$ , since  $\nu \approx 1.1 \times 10^{15}$  Hz. In order to operate Al<sup>+</sup> as a frequency standard with fractional frequency uncertainty below  $6 \times 10^{-17}$ , the blackbody shift must be calibrated experimentally.

### C. Near-infrared Stark shift measurement

Ideally, we would measure the shift of the clock transition due to a known intensity of 10  $\mu$ m radiation, since the room temperature blackbody field is centered at this wavelength. However, the windows of our experimental apparatus are opaque to wavelengths longer than 3  $\mu$ m. Instead, we measure the Stark shift due to near-infrared radiation, and use this measurement to estimate the blackbody shift.

The output of a fiber laser (600 mW with  $\pm 200$  mW fluctuations) at 1126 nm was focussed onto an Al<sup>+</sup> ion, and switched on and off at regular intervals. A stable ULE reference cavity was simultaneously locked to the <sup>1</sup>S<sub>0</sub>  $\rightarrow$  <sup>3</sup>P<sub>0</sub> transition, via an acousto-optic frequency shifter, and the frequency shift due to the Stark shifting beam was tracked and recorded. These measurements were repeated for various lateral (x,y) displacements of the Stark shifting beam, as shown in Figure 1, in order to estimate the beam waist ( $w_0 = 100 \pm 10$   $\mu$ m).

The resulting differential polarizability is  $\Delta\alpha(2\pi c/(1126 \text{ nm})) = 4\pi\epsilon_0 \times (1.6 \pm 0.5) \times 10^{-31}$  m<sup>3</sup>, limited in accuracy by power fluctuations of the Stark shifting laser.

### D. Extrapolation to zero frequency

The following relates this measurement to the differential polarizability at 0 Hz, by expanding Eq. (1) in small parameters. Two facts specific to Al<sup>+</sup> are used.

1. All strong transitions connecting to either clock state are in the deep UV ( $\lambda < 186$  nm).
2. The strongest transitions contributing to the sum in Eq. (1) are near each other ( $\lambda \approx 170$  nm).

Let  $\delta_i \equiv (\omega/\omega_i)^2$ . For the <sup>1</sup>S<sub>0</sub> and <sup>3</sup>P<sub>0</sub> states in Al<sup>+</sup>,  $\delta_i < 0.03$ , when  $\omega = 2\pi c/(1126 \text{ nm})$ . Expanding

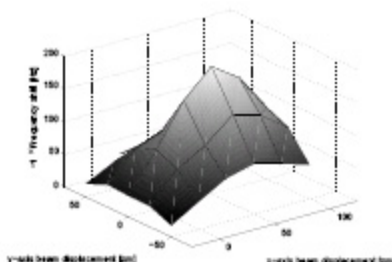


FIG. 1: Clock transition Stark shift vs. beam position. A Gaussian beam profile fit yields  $w_0 = 100$   $\mu$ m for the beam waist, with a peak shift of -190 Hz.

Eq. (1) in powers of  $\delta_i$  yields

$$\alpha(\omega) = \alpha(0) + \frac{e^2}{m_e} \sum_i \frac{f_i}{\omega_i^2} (\delta_i + \delta_i^2 + \dots). \quad (3)$$

Thus, the differential polarizability between the <sup>1</sup>S<sub>0</sub> and <sup>3</sup>P<sub>0</sub> states is

$$\Delta\alpha(\omega) = \Delta\alpha(0) + \frac{e^2}{m_e} \sum_i \frac{f_i}{\omega_i^2} (\delta_i + \delta_i^2 + \dots), \quad (4)$$

where we sum over all transitions connecting to the <sup>1</sup>S<sub>0</sub> and <sup>3</sup>P<sub>0</sub> states. Positive oscillator strengths are used for the transitions connecting to <sup>3</sup>P<sub>0</sub>, and negative oscillator strengths are used for the <sup>1</sup>S<sub>0</sub> transitions.

Now let  $\delta_0 \equiv (\omega/\omega_0)^2$ , where  $\omega_0 = 2\pi c/(171 \text{ nm})$ , and let  $\epsilon_i \equiv \delta_i - \delta_0$ . This value of  $\delta_0$  is chosen because the strong transitions all lie near 171 nm. Then

$$\Delta\alpha(0) = \frac{\Delta\alpha(\omega) - \frac{e^2}{m_e} \sum_i \frac{f_i}{\omega_i^2} (\epsilon_i + \delta_i^2 + \dots)}{1 + \delta_0}. \quad (5)$$

All of the terms after the summation sign are small, as can be seen in Table II. For the strongest transitions  $\epsilon_i$  is small, because all strong transitions are near 171 nm. For the weaker transitions  $f_i/\omega_i^2$  is small. To test the merits of this estimate, we propagate the uncertainties  $\sigma_{f_i}$  (see Table II) in the various  $f_i$  via Eq. (5), which results in an uncertainty in  $\Delta\alpha(0)$  of

$$\sigma_{\Delta\alpha(0)} = \frac{e^2/m_e}{1 + \delta_0} \sqrt{\sum_i \left[ \frac{\sigma_{f_i}}{\omega_i^2} (\epsilon_i + \delta_i^2 + \dots) \right]^2}. \quad (6)$$

Note that our choice of  $\delta_0$  minimizes the uncertainty  $\sigma_{\Delta\alpha(0)}$ . Numerically we find  $\sigma_{\Delta\alpha(0)} = 4\pi\epsilon_0 \times 1.7 \times 10^{-33} \text{ m}^3 \approx 0.01 \times \Delta\alpha(2\pi c/(1126 \text{ nm}))$ . Thus,  $\Delta\alpha(0)$  can be deduced from our measurement of  $\Delta\alpha(\omega)$  at 1126 nm with an additional uncertainty of 1 %. Eq. (5) yields  $\Delta\alpha(0) = 4\pi\epsilon_0 \times (1.5 \pm 0.5) \times 10^{-31} \text{ m}^3$ .

### E. Estimate of blackbody shift

Since the frequency of blackbody radiation (centered at 10  $\mu\text{m}$  wavelength) is closer to 0 Hz than the frequency of the applied 1126 nm radiation, we expect to relate  $\Delta\alpha(0)$  to  $\Delta\alpha(2\pi c/(10 \mu\text{m}))$  with even less uncertainty than our estimate of  $\Delta\alpha(0)$  from  $\Delta\alpha(2\pi c/(1126 \text{ nm}))$ . As before, we can propagate the errors  $\sigma_{f_i}$  through the result. The calculation follows from Section D and Eq. (2), and we simply write the room temperature result as

$$\Delta\nu = -\frac{\pi k_B^4 T^4}{60\epsilon_0 \hbar^4 c^3} (\Delta\alpha(0) \times 1.00024), \quad (7)$$

or numerically,  $\Delta\nu = -0.008(3) \text{ Hz}$ .

### F. Conclusion

We have measured the differential polarizability of the  $^{27}\text{Al}^+ \ ^1\text{S}_0 \rightarrow \ ^3\text{P}_0$  clock transition at 1126 nm. We have also found expressions relating the differential polarizabilities at various drive frequencies, in which the effect of uncertainties in the oscillator strengths is minimized. In particular,  $\Delta\alpha(0)$  is found from  $\Delta\alpha(2\pi c/(1126 \text{ nm}))$  with 1 % added fractional uncertainty, while allowing conservative uncertainties of 20 % or larger in the oscillator strengths. From  $\Delta\alpha(0)$  we calculate the blackbody shift with negligible added uncertainty. The fractional room temperature blackbody shift  $\Delta\nu/\nu = (-8 \pm 3) \times 10^{-18}$  is substantially lower for the  $^{27}\text{Al}^+ \ ^1\text{S}_0 \rightarrow \ ^3\text{P}_0$  transition than for other atomic frequency standards currently under development (see Table I). The uncertainty in this value could be lowered substantially by improving the power stability of the 1126 nm Stark shifting laser.

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TABLE II: Transition wavelengths ( $\lambda_i$ ) and oscillator strengths ( $f_i$ ) used to estimate  $\Delta\alpha(0)$ . Transitions are in descending order of  $f_i$  magnitude. Negative oscillator strengths are used for transitions connecting to the  $3s^2\ ^1S_0$  ground state. Fractional uncertainties  $\sigma_{f_i}/|f_i|$  were taken from the NIST Atomic Spectra Database [9] where available, and doubled. Where no uncertainty is available, a fractional uncertainty of 1 is assumed.  $\delta_i = (\omega/\omega_i)^2$  where  $\omega = 2\pi c/(1126\text{ nm})$ ,  $\omega_i = 2\pi c/\lambda_i$ , and  $\epsilon_i = \delta_i - (171/1126)^2$ . The sixth column lists the summands of Eq. (5) truncated after  $\delta_i^2$ .

$f_i$	$\sigma_{f_i}/ f_i $	$\lambda_i$ [nm]	$\delta_i$	$\epsilon_i$	$\frac{e^2}{m_e} \frac{f_i}{\omega_i^2} (\epsilon_i + \delta_i^2 + \delta_i^3)$ [ $4\pi\epsilon_0 \times \text{nm}^3 \times 10^{-33}$ ]	from	to	ref.
-1.830000	0.20	167.079	0.0220	-0.0010	2.005625	3s2 1S0	3s3p 1P1	[9]
0.903000	0.20	171.944	0.0233	0.0003	1.546615	3s3p 3P0	3s3d 3D1	[9]
0.612000	0.50	176.198	0.0245	0.0014	2.762716	3s3p 3P0	3p2 3P1	[9]
0.129000	0.20	185.593	0.0272	0.0041	1.541754	3s3p 3P0	3s4s 3S1	[9]
0.059000	1.00	118.919	0.0112	-0.0119	-0.701593	3s3p 3P0	3s4d 3D1	[9]
0.018000	1.00	104.789	0.0087	-0.0144	-0.202074	3s3p 3P0	3s5d 3D1	[9]
0.016556	1.00	120.919	0.0115	-0.0115	-0.196866	3s3p 3P0	3s5s 3S1	[10]
0.005922	1.00	105.460	0.0088	-0.0143	-0.066807	3s3p 3P0	3s6s 3S1	[10]
0.004078	1.00	98.598	0.0077	-0.0154	-0.043385	3s3p 3P0	3s6d 3D1	[10]
-0.003020	1.00	93.527	0.0069	-0.0162	0.030381	3s2 1S0	3s4p 1P1	[10]
0.002889	1.00	98.905	0.0077	-0.0153	-0.030830	3s3p 3P0	3s7s 3S1	[10]
0.001889	1.00	95.263	0.0072	-0.0159	-0.019393	3s3p 3P0	3s7d 3D1	[10]
0.001656	1.00	95.429	0.0072	-0.0159	-0.017030	3s3p 3P0	3s8s 3S1	[10]
-0.001100	1.00	71.470	0.0040	-0.0190	0.007625	3s2 1S0	3s7p 1P1	[10]
-0.001090	1.00	74.118	0.0043	-0.0187	0.007995	3s2 1S0	3s6p 1P1	[10]
-0.001050	1.00	69.949	0.0039	-0.0192	0.007035	3s2 1S0	3s8p 1P1	[10]
0.001050	1.00	93.341	0.0069	-0.0162	-0.010539	3s3p 3P0	3s9s 3S1	[10]
0.001019	1.00	93.241	0.0069	-0.0162	-0.010214	3s3p 3P0	3s8d 3D1	[10]
-0.000998	1.00	68.994	0.0038	-0.0193	0.006541	3s2 1S0	3s9p 1P1	[10]
-0.000948	1.00	68.353	0.0037	-0.0194	0.006120	3s2 1S0	3s10p 1P1	[10]
-0.000858	1.00	67.902	0.0036	-0.0194	0.005480	3s2 1S0	3s11p 1P1	[10]
0.000708	1.00	91.980	0.0067	-0.0164	-0.006985	3s3p 3P0	3s10s 3S1	[10]
0.000613	1.00	91.916	0.0067	-0.0164	-0.006048	3s3p 3P0	3s9d 3D1	[10]
-0.000567	1.00	79.448	0.0050	-0.0181	0.004612	3s2 1S0	3s5p 1P1	[10]
0.000501	1.00	91.041	0.0065	-0.0165	-0.004885	3s3p 3P0	3s11s 3S1	[10]
0.000398	1.00	90.997	0.0065	-0.0165	-0.003876	3s3p 3P0	3s10d 3D1	[10]
0.000124	1.00	70.040	0.0039	-0.0192	-0.000835	3s3p 3P0	3p5p 3P1	[10]
0.000116	1.00	65.800	0.0034	-0.0196	-0.000701	3s3p 3P0	3p6p 3P1	[10]
0.000083	1.00	63.690	0.0032	-0.0199	-0.000477	3s3p 3P0	3p7p 3P1	[10]
0.000059	1.00	62.480	0.0031	-0.0200	-0.000328	3s3p 3P0	3p8p 3P1	[10]
0.000043	1.00	61.710	0.0030	-0.0201	-0.000233	3s3p 3P0	3p9p 3P1	[10]
0.000032	1.00	61.190	0.0030	-0.0201	-0.000170	3s3p 3P0	3p10p 3P1	[10]
0.000001	1.00	80.860	0.0052	-0.0179	-0.000009	3s3p 3P0	3p4p 3P1	[10]
SUM					6.609223			
UNCERTAINTY					1.685575			