Atomic clocks with suppressed blackbody radiation shift

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We develop a nonstandard concept of atomic clocks where the blackbody radiation shift (BBRS) and its temperature fluctuations can be dramatically suppressed (by one to three orders of magnitude) independent of the environmental temperature. The suppression is based on the fact that in a system with two accessible clock transitions (with frequencies $\nu_1$ and $\nu_2$) which are exposed to the same thermal environment, there exists a “synthetic” frequency $\nu_{\text{syn}} \propto (\nu_1 - \varepsilon_1 \nu_2)$ largely immune to the BBRS. As an example, it is shown that in the case of $^{171}$Yb$^+$ it is possible to create a clock in which the BBRS can be suppressed to the fractional level of $10^{-18}$ in a broad interval near room temperature ($300\pm15$ K). We also propose a realization of our method with the use of an optical frequency comb generator stabilized to both frequencies and its temperature fluctuations can be dramatically suppressed (by one to three orders of magnitude) independent of the environmental temperature. The suppression is based on the fact that for the large majority of transitions in atoms or ions that are of interest in atomic frequency standards by one to three orders of magnitude without using cryogenic techniques and precise temperature stabilization. Our approach is based on the use of two reference transitions in an identical thermal environment. We show that in such a system there exists a combined frequency for which the BBRS is significantly suppressed over a wide temperature range. For instance, a trapped $^{171}$Yb$^+$ ion meets this condition in a straightforward way, because $^{171}$Yb$^+$ has at least three suitable reference transitions: an electric-quadrupole and an electric-octupole optical transition and a magnetic-dipole radiofrequency (rf) transition between the ground-state hyperfine sublevels. Apart from laboratory standards, the proposed method can be particularly useful in cases where it is impossible to control the environmental temperature with sufficient accuracy or to use cryogenic techniques, for instance in transportable frequency standards or in space-based clocks that approach the Sun in order to test the local position invariance underlying General Relativity.

Our approach is based on the fact that for the large majority of transitions in atoms or ions that are of interest as frequency standard reference transitions, the temperature dependence $\Delta(T)$ of the BBRS is very well approx-
imated by the law $\propto T^4$. Consider now two clock transitions with frequencies $\nu_1^{(0)}$ and $\nu_2^{(0)}$ exposed to the same thermal environment, i.e., located in the same probe volume. We assume that $\nu_1^{(0)} < \nu_2^{(0)}$. The effect of the BBRS on each transition frequency can be represented as:

$$\nu_j(T) \approx \nu_j^{(0)} + a_j \left( \frac{T}{T_0} \right)^4 \quad (j = 1, 2), \quad (1)$$

where $a_j$ is an individual characteristic of the transition $j$ determined by the atomic structure and $T_0$ is the mean temperature of the clock operation. Let us introduce the coefficient $\varepsilon_{12} = a_1/a_2$. As is easily seen, the following superposition does not experience the BBRS: $\nu_1(T) - \varepsilon_{12} \nu_2(T) = \nu_1^{(0)} - \varepsilon_{12} \nu_2^{(0)}$. In compliance with this we define a new “synthetic” frequency $\nu_{\text{syn}}$ as

$$\nu_{\text{syn}} = R[\nu_1(T) - \varepsilon_{12} \nu_2(T)] = R[\nu_1^{(0)} - \varepsilon_{12} \nu_2^{(0)}], \quad (2)$$

where $R$ is some numerical multiplier whose value can be chosen freely. Thus, one can use the frequency $\nu_{\text{syn}}$ as a new clock output frequency which is immune to the BBRS and to fluctuations in the operating temperature, while the thermal shifts $a_j T^4$ of the working frequencies $\nu_j$ can be large.

One possibility is to independently measure both frequencies $\nu_{1,2}(T)$ and to use for the clock operation the synthetic frequency according to Eq. (2) (assuming, for example, $R = \pm 1$). In this case, obviously the synthetic frequency does not directly correspond to any frequency of a physical signal. Another approach is to synthesize this frequency as a real physical signal by means of an optical frequency comb generator. Let us consider the situation where two modes of the frequency comb generator are stabilized to the two optical frequencies $\nu_{1,2}(T) = f_0 + n_{1,2} f_r$ at a given temperature $T$ (see Fig. 1). As a result, the parameters of the comb spectrum, i.e., the pulse repetition rate $f_r$ and the offset frequency $f_0$ are unambiguously determined and the frequency of the $m$-th mode equals:

$$\nu_m(T) = f_0 + m f_r = \frac{m (\nu_2^{(0)} - \nu_1^{(0)}) + n_2 \nu_1^{(0)} - n_1 \nu_2^{(0)}}{n_2 - n_1} + \frac{m(a_2 - a_1) + n_2 a_1 - n_1 a_2}{n_2 - n_1} \left( \frac{T}{T_0} \right)^4. \quad (3)$$

From this expression one can define a number $m = m_0$ for which the coefficient of the temperature-dependent term is zero:

$$m_0 = \frac{n_1 a_2 - n_2 a_1}{a_2 - a_1} = \frac{n_1 - \varepsilon_{12} n_2}{1 - \varepsilon_{12}}. \quad (4)$$

This shows that the BBRS is suppressed for the frequency $\nu_{m_0}$. After a simple transformation we see that the frequency $\nu_{m_0}$ is the synthetic frequency defined in Eq. (2),

$$\nu_{m_0} = \nu_{\text{syn}} = \frac{\nu_1^{(0)} - \varepsilon_{12} \nu_2^{(0)}}{1 - \varepsilon_{12}}, \quad (5)$$

as it should be. Here, $m_0$ is the natural number closest to the value of the right-hand side of Eq. (4). For this it is necessary to satisfy the condition $m_0 > 0$ that is equivalent to $\nu_{\text{syn}} > 0$ in Eq. (5).

Apart from the frequency $\nu_{m_0}$, which is a component of the optical spectrum of the frequency comb generator, in our system one can also define the much smaller frequency

$$\frac{\nu_{m_0}}{m_0} = f_r + \frac{f_0}{m_0} \quad (6)$$

which corresponds to a rf standard at $\nu_{m_0}/m_0$. Since the frequencies $f_r$ and $f_0$ can be extracted from a stabilized comb generator with negligible error, one can use them to synthesize $\nu_{m_0}/m_0$. This synthesized radiofrequency has the same immunity to BBRS as $\nu_{m_0}$. It is interesting to note that the radiofrequency given in Eq. (6) is well-defined in our system even if $m_0 < 0$ in Eq. (4), i.e., if the basic frequency component $\nu_{m_0}$ exists only virtually.

As was shown above, in the case of a frequency comb stabilized to two BBR-shifted clock transitions with frequencies $\nu_1(T)$ and $\nu_2(T)$, there exists a frequency component $\nu_{m_0}$ (for $m_0 > 0$) for which the thermal shift and the sensitivity to temperature fluctuations vanish. This frequency component can serve as an atomic frequency standard. In practice, the BBRS is strongly suppressed for a range of comb frequencies around $\nu_{m_0}$. The residual shift of frequency components $\nu_{m_0 \pm l}$ near $\nu_{m_0}$ is given by:

$$\nu_{m_0 \pm l} = \nu_{m_0} \pm l f_r = \nu_{m_0} \pm \frac{\nu_2^{(0)} - \nu_1^{(0)}}{n_2 - n_1} \pm \frac{a_2 - a_1}{n_2 - n_1} \left( \frac{T}{T_0} \right)^4. \quad (7)$$

This indicates that the suppression is effective as long as $(n_2 - n_1) \gg l$. For example, for frequencies $\nu_1^{(0)}$ and $\nu_2^{(0)}$ in the optical range, the comb mode index difference

**FIG. 1:** Illustration of femtosecond comb stabilized to the two clock transitions with frequencies $\nu_1$ and $\nu_2$ at a given temperature $T$.
\((n_2 - n_1)\) will typically be in the range of \(10^5\) or higher (see the discussion for the case of \(^{171}\text{Yb}^+\) below). On the whole, the choice of the exact value of the synthetic frequency \(\nu_{\text{syn}}\) is to some extent arbitrary if one takes into account that the coefficient \(\varepsilon_{12}\) is only known with limited accuracy and that Eq. (1) is an approximation that neglects higher-order terms in the temperature dependence of the BBRS [18]. Including higher-order terms the BBRS can be expressed as:

\[
\Delta^{(j)}(T) = a_j \left( \frac{T}{T_0} \right)^4 + b_j \left( \frac{T}{T_0} \right)^6 + \ldots \quad (j = 1, 2). \quad (8)
\]

From this we can estimate a basic limitation of the possibility to suppress the BBRS and its temperature dependence. Usually, near room temperature \(T_0 = 300\ \text{K}\) the contribution of the higher terms \([b_j(T/T_0)^6 + \ldots]\) is a factor of 10 to 100 smaller than that of the main \(T^4\)-term [19]. This indicates that here it would not be useful to suppress the \(T^4\)-dependence of \(\nu_{\text{syn}}\) to better than one to three orders of magnitude because higher-order contributions to the BBRS remain uncompensated. For example, in order to achieve a suppression factor of \(10^6\) for the \(T^4\)-dependence, it would be sufficient to know the coefficient \(\varepsilon_{12}\) with relative uncertainty of \(10^{-2}\).

It may be noted that apart from theoretical calculations the coefficient \(\varepsilon_{12}\) can be determined by purely experimental means. To do this we can apply a quasistatic electric field (or the field of an infrared laser) to determine the involved atomic energy levels. From a practical point of view it is very advantageous that we do not need to know the magnitude of the electric field at the place of the atoms because we only have to determine the ratio \(a_1/a_2\). If a frequency comb generator is stabilized to \(\nu_1\) and \(\nu_2\) as shown in Fig.1, the frequency \(\nu_{\text{syn}}\) can be identified in a direct way as the frequency component which does not experience any scalar Stark shift in the applied quasistatic field.

As an example that permits the practical realization of the ideas presented above, we consider the ion \(^{171}\text{Yb}^+\). As shown in Fig.2 the level system of \(^{171}\text{Yb}^+\) provides two narrow-linewidth transitions from the ground state in the visible spectral range which can be used as reference transitions of an optical frequency standard: the quadrupole transition \(^2S_{1/2}(F = 0) \rightarrow ^2D_{3/2}(F = 2)\), \(\lambda \approx 436\ \text{nm}\) and the octupole transition \(^2S_{1/2}(F = 0) \rightarrow ^2F_{7/2}(F = 3)\), \(\lambda \approx 467\ \text{nm}\). More detailed information on the spectroscopy of these transitions can be found in [15, 17, 20]. It may be noted that the case of \(^{171}\text{Yb}^+\) is especially attractive because here both clock transitions lie in a technically convenient frequency range and experience exactly the same thermal environment if probed in one ion.

The BBRS of the quadrupole and octupole transitions of \(^{171}\text{Yb}^+\) were calculated in Ref. [21]. This calculation is based on calculated oscillator strengths and experimental lifetime and polarizability data. The room-temperature BBRS of the quadrupole transition is calculated as \(a_{\text{quad}} = -0.35(7)\ \text{Hz}\) (fractional shift 5.1(1.1) \times 10^{-16}) and that of the octupole transition as \(a_{\text{oct}} = -0.15(7)\ \text{Hz}\) (fractional shift 2.4(1.1) \times 10^{-16}). The relatively small value of \(a_{\text{oct}}\) and the large relative uncertainty is due to nearly equal shifts of the \(^2S_{1/2}\) and \(^2F_{7/2}\) levels.

Using the results of Ref. [21], for \(^{171}\text{Yb}^+\) we find that \(\varepsilon_{12} = a_{\text{oct}}/a_{\text{quad}} = 0.43(22)\). We expect that the large uncertainty of this value can be reduced to less than 1% by improved atomic structure calculations or by a direct measurement of \(\varepsilon_{12}\) as discussed above. In the following, we will not take into account the present uncertainty because our conclusions remain qualitatively unchanged for all values of \(\varepsilon_{12}\) in this uncertainty range. In particular, we find the synthetic frequency \(\nu_{\text{syn}}^{(\text{comb})} = 607\ \text{THz}\), corresponding to a wavelength \(\lambda_{\text{syn}} \approx 494\ \text{nm}\). This frequency lies sufficiently close to the initial reference transitions at 436 nm and 467 nm that it can be generated as a spectral component of a femtosecond comb generator that is locked to the reference transitions as shown in Fig. 1.

The higher-order contributions in Eq. (8) to the BBRS of the octupole reference transition are negligible compared to that of the quadrupole transition. For the latter, we find \(b/a \approx 0.1\) at \(T_0 = 300\ \text{K}\). As a result, we estimate that the BBRS can be suppressed to the fractional level of \(2.7 \times 10^{-17}\) at 300 K with variations at the level of \(\pm 5 \times 10^{-18}\) if the ambient temperature varies in a broad interval of \(\pm 15\ \text{K}\).

It is also possible to estimate the frequency interval around \(\nu_{\text{syn}}^{(\text{comb})}\) where the components of the comb spectrum have a similar level of suppression of the thermal shift and of its fluctuations. For a suppression factor of \(10^2\), this interval has a width of the order of 1000 GHz. Thus, for \(d \approx 100\ \text{MHz}\), the indicated interval contains \(10^4\) comb modes, each of which could be used as a stable frequency reference.

FIG. 2: (Color online) Section of the energy level scheme of \(^{171}\text{Yb}^+\), showing the hyperfine levels of the \(^2S_{1/2}\) ground state and the two lowest-lying excited states, which are metastable. Hyperfine splittings are not drawn to scale.
Other variants of BBRS-free optical frequency standards at a synthetic frequency can be conceived based on transitions $^1S_0 \rightarrow ^3P_0$ in alkaline-earth-like neutral atoms confined in an optical lattice. Consider, for instance, the combination of the reference transitions of strontium ($\nu_1 \approx 429$ THz, $\lambda \approx 698$ nm) and ytterbium ($\nu_2 \approx 518$ THz, $\lambda \approx 578$ nm). Using the calculations in Ref. 22, in this case we obtain $\varepsilon_{12} \approx 1.69$ and an estimated synthetic frequency $\nu_{\text{syn}}^{(\text{comb})} \approx 648$ THz ($\lambda_{\text{syn}} \approx 463$ nm). The technical realization of this variant requires the operation of two lattice-based clocks with different atoms (Sr and Yb) in the same vacuum chamber.

So far we have considered examples where both reference transition frequencies lie in the optical region. However, it is also possible to realize schemes where the high frequency $\nu_2$ is optical, but the lower frequency $\nu_1$ corresponds to a fine- or hyperfine-structure splitting so that it lies in the terahertz or microwave range. In contrast to the above example of ion Yb$^+$, which seems unique because it provides two optical reference transitions, in this case one can find many appropriate schemes that use a single atomic species. Also the ion $^{171}$Yb$^+$ offers the possibility of using the ground-state hyperfine transition $F = 0 \rightarrow F = 1$ at $\nu_1 \approx 12.6$ GHz (see Fig. 2) as a low-frequency reference transition. For the combination with the octupole transition $^2S_{1/2}(F = 0) \rightarrow ^2F_{7/2}(F = 3)$ at $\nu_2 \approx 642$ THz, a numerical estimate yields $\varepsilon_{12} \approx 6.6 \times 10^{-5}$ and a synthetic frequency $\nu_{\text{syn}} = - (\nu_1 - \varepsilon_{12} \nu_2) \approx 30$ GHz. (We expect that our estimate on $\varepsilon_{12}$ is accurate to $\pm 20\%$, but it is not principal for further results.) Here the $T^6$-contribution to the BBRS (see Eq. (8)) limits the BBRS suppression at the fractional level of $7.5 \times 10^{-19}$ at $T=300$ K with variations of $\pm 2 \times 10^{-19}$ in the temperature interval of $300 \pm 15$ K. However, we should also take into account the shift of the ground-state hyperfine levels ($\propto T^2$) due to the magnetic blackbody radiation field $\mathcal{E}$. The corresponding BBRS of the hyperfine frequency $F = 0 \rightarrow F = 1$ for $^{171}$Yb$^+$ is:

$$\Delta_{\text{magn}}^{(1)}(T) = -1.616 \times 10^{-7} \times \left( \frac{T(K)}{300} \right)^2 \text{Hz}. \quad (9)$$

For $\nu_{\text{syn}} = 30$ GHz this shift results in a fractional level of $5.4 \times 10^{-18}$ at $T=300$ K with a variation of $\pm 5 \times 10^{-19}$ for $300 \pm 15$ K. Since the magnetic BBRS contribution can be readily calculated with an accuracy of less than $1\%$, it is possible to apply a corresponding correction to $\nu_{\text{syn}}$ with an uncertainty contribution of less than $10^{-19}$.

For $^{171}$Yb$^+$ we have thus shown the possibility to create a synthetic-frequency-based atomic clock with a fractional uncertainty contribution due to BBRS of $\approx 1.5 \times 10^{-18}$ in a broad interval of $300 \pm 15$ K. To achieve this, we need to know the coefficient $\varepsilon_{12}$ with a relative accuracy in the range of 0.1–0.2\%. In order to reduce the BBRS uncertainty contribution to less than $10^{-17}$, the value of $\varepsilon_{12}$ needs only be known with a relative accuracy of $3\%$. We also have pointed out that for $^{171}$Yb$^+$ the combination of the octupole optical clock transition with the ground-state hyperfine transition can yield a much better BBRS suppression than the combination of the octupole and quadrupole optical clock transitions. The use of the quadrupole transition yields a lower BBRS suppression because the upper level $^2D_{3/2}$ is connected to the $^2P_{1/2}$ level by a strong infrared transition at 2.44 $\mu$m, which produces a relatively large $T^6$-contribution to the BBRS. The final comparison of the two options for BBRS suppression should also include detailed estimates on the magnitudes of other systematic uncertainty contributions in the considered experimental setup.

The concept of a synthetic atomic frequency standard based on two reference transitions can also be extended to the case that both reference frequencies $\nu_{1,2}$ lie in the microwave range. Atomic fountain clocks are based on reference transitions in the microwave range between the ground-state hyperfine sublevels of alkali atoms. For a synthetic atomic fountain frequency standard, for instance the combination $^{87}$Rb ($\nu_1 \approx 6.8$ GHz) and $^{133}$Cs ($\nu_2 \approx 9.2$ GHz) can be considered. Here, at the synthetic frequency ($\nu_1 - \varepsilon_{12} \nu_2$) $\approx 1.9$ GHz it is possible to suppress the fractional BBRS of the individual standards by two orders of magnitude. It is interesting to note that nearly optimal conditions for the efficient suppression of the BBRS are realized in the dual Rb/Cs fountain clock described in Ref. 22 because here both reference transitions are exposed to the same thermal environment.

We finally note that the $^{171}$Yb$^+$ optical frequency standard is a very sensitive system for a search for temporal variations of the fine structure constant $\alpha$. The frequencies of the quadrupole and octupole reference transitions of Yb$^+$ have significant contributions from relativistic effects and would undergo changes with different sign in consequence of a change of $\alpha$. The synthetic frequency that eliminates the BBRS retains this sensitivity. The $\alpha$-dependence of an atomic transition frequency may be expressed generally as $\nu = \nu_0 + q \alpha$, where $x \equiv (\alpha/\alpha_0)^2 - 1$, $\nu_0$ defines the frequency at the present value of the fine structure constant, $\alpha_0$, and $q$ quantifies the sensitivity to changes of $\alpha$. The parameter for the synthetic frequency is simply given by $\nu_{\text{syn}} = (\nu_0 + \varepsilon_{12} \nu_2)/(1 - \varepsilon_{12})$. For Yb$^+$, with $q$ parameters as given in Ref. 24, $\nu_{\text{syn}}$ amounts to about $-3220$ THz. Comparison with the Yb$^+$ synthetic frequency $\nu_{\text{syn}}^{(\text{comb})} \approx 607$ THz indicates the strong sensitivity. In a test for variations of $\alpha$, the synthetic frequency would have to be compared to an “anchor” reference transition with small $q$ value, like the $^1S_0 \rightarrow ^3P_0$ transition in Al$^+$. In conclusion, we have proposed and developed the concept of an atomic frequency standard where the frequency shift due to the ambient blackbody radiation and related fluctuations of the output frequency can be suppressed by one to three orders of magnitude without us-
ing cryogenic techniques. We also expect that our results will stimulate refined atomic structure calculations on Yb\(^+\) and other atomic systems that are of interest in this context. Such calculations can yield precise values for the frequency synthesis parameter \(\varepsilon_{12}\) and determine limitations of the achievable BBRS suppression.

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[1] T. Rosenband et al., Science 319, 1808 (2008).
[2] T. Akatsuka, M. Takamoto, and H. Katori, Nature Physics 4, 954 (2008).
[3] A. D. Ludlow et al., Science 319, 1805 (2008).
[4] S. G. Turyshev et al., in From Quantum to Cosmos: Fundamental Physics Research in Space (World Scientific, Singapore, 2009).
[5] Atomic Clocks and Fundamental Constants, Eur. Phys. J. ST 163 (2008).
[6] W. M. Itano, L. L. Lewis, and D. J. Wineland, Phys. Rev. A 25, 1233(R) (1982).
[7] W. H. Oskay et al., Phys. Rev. Lett. 97, 020801 (2006).
[8] F. Levi et al., IEEE Trans. UFFC 57, 600 (2010).
[9] T. Middelmann et al., arXiv:1009.2017 (2010).
[10] E. J. Angstmann, V. A. Dzuba, and V. V. Flambaum, Phys. Rev. A 74, 023405 (2006).
[11] M. S. Safronova et al., IEEE Trans. UFFC 57, 94 (2010).
[12] C. W. Chou, D. B. Hume, J. C. J. Koelmeij, D. J. Wineland, and T. Rosenband, Phys. Rev. Lett. 104, 070802 (2010).
[13] Th. Becker et al., Phys. Rev. A 63, 051801(R) (2001).
[14] The fractional room-temperature BBRS of the In\(^+\) reference transition has been calculated as \(1.4(1) \times 10^{-17}\) by M. G. Kozlov and M. S. Safronova (private communication).
[15] Chr. Tamm, S. Weyers, B. Lipphardt, and E. Peik, Phys. Rev. A 80, 043403 (2009).
[16] K. Hosaka, S. A. Webster, A. Stannard, B. R. Walton, H. S. Margolis, and P. Gill, Phys. Rev. A 79, 033403 (2009).
[17] I. Sherstov, M. Okhapkin, B. Lipphardt, Chr. Tamm, and E. Peik, Phys. Rev. A 81, 021805(R) (2010).
[18] J. W. Farley, W. H. Wing, Phys. Rev. A 23, 2397 (1981).
[19] V. G. Pal’chikov, Yu. S. Domnin, and A. V. Novoselov, J. Opt. B: Quantum Semiclass. Opt. 5, 131 (2003).
[20] T. Schneider, E. Peik, and Chr. Tamm, Phys. Rev. Lett. 94, 230801 (2005).
[21] S. N. Lea, S. A. Webster, and G. P. Barwood, in Proceedings of the 20th European Frequency and Time Forum, Braunschweig, 2006 (PTB, Braunschweig, Germany, 2006), p. 302.
[22] S. G. Porsev and A. Derevianko, Phys. Rev. A 74, 020502 (2006).
[23] J. Guéna et al., IEEE Trans. UFFC 57, 647 (2010).
[24] V. A. Dzuba, V. V. Flambaum, and M. V. Marchenko, Phys. Rev A 68, 022506 (2003).
[25] S. N. Lea, Rep. Progr. Phys. 70, 1473 (2007).