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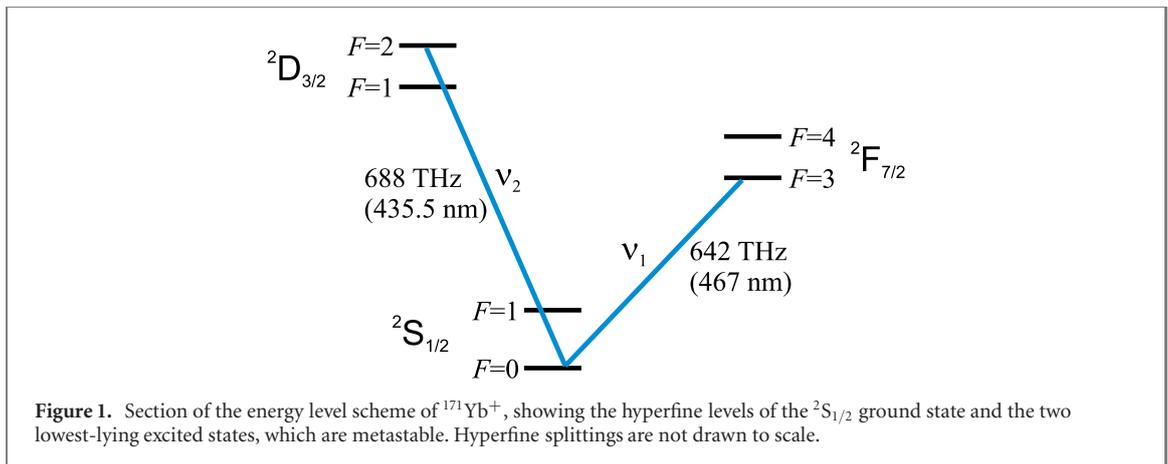
Keywords: ultra-precise atomic clock, BBR shift, temperature control

## Abstract

We develop a method of synthetic frequency generation to construct an atomic clock with blackbody radiation (BBR) shift uncertainties below  $10^{-19}$  at environmental conditions with a very low level of temperature control. The proposed method can be implemented for atoms and ions, which have two different clock transitions with frequencies  $\nu_1$  and  $\nu_2$  allowing to form a synthetic reference frequency  $\nu_{\text{syn}} = (\nu_1 - \varepsilon\nu_2)/(1 - \varepsilon)$ , which is absent in the spectrum of the involved atoms or ions. Calibration coefficient  $\varepsilon$  can be chosen such that the temperature dependence of the BBR shift for the synthetic frequency  $\nu_{\text{syn}}$  has a local extremum at an arbitrary operating temperature  $T_0$ . This leads to a weak sensitivity of BBR shift with respect to the temperature variations near operating temperature  $T_0$ . As a specific example, the  $\text{Yb}^+$  ion is studied in detail, where the utilized optical clock transitions are of electric quadrupole ( $S \rightarrow D$ ) and octupole ( $S \rightarrow F$ ) type. In this case, temperature variations of  $\pm 7$  K lead to BBR shift uncertainties of less than  $10^{-19}$ , showing the possibility to construct ultra-precise combined atomic clocks (including portable ones) without the use of cryogenic techniques.

Historically, the developments in timekeeping have always led to most precise scientific instruments. At present, atomic clocks rightfully hold the leading position and are of exceptional importance both in the field of fundamental science and for numerous practical applications such as the global positioning systems and relativistic geodesy [1–5]. Supported by high-precision laser-spectroscopy and the advanced degree of control of trapped atoms and ions, these table-top experiments achieve fractional instabilities of  $10^{-18}$  in atomic transition frequency determination [6–8]. This opens up the competitive search for new physics, e.g. dark matter and the variation of the fine structure constant [9], a potential fifth force [10] and test of general relativity [11, 12] to understand the discrepancy between reality and the predictions made by the standard model of particle physics. Today's work on optical clocks targets the low  $10^{-19}$  level, allowing for even more precise tests of the underlying fundamental theories.

A main problem in ultra-precision atomic clocks is the blackbody radiation (BBR) shift of the clock transition, caused by the interaction of the atoms (ions) with thermal photons that make up the thermodynamic environment [13, 14]. It is this fundamental shift and its variations that are a major obstacle to reach the  $10^{-19}$  level. Indeed, in most cases the BBR shift at room temperature  $T \sim 300$  K has a relative magnitude of  $10^{-15}$ – $10^{-16}$  with respect to the frequency of the reference optical transition [15] (with some exceptions [16–21]). Therefore, in order that the long-term variations in BBR shift does not exceed  $10^{-19}$ , a high level of thermostabilization of the experimental setup itself and its thermodynamic environment is required, which is a complex technical problem. Thus, a number of laboratories have taken the path of using cryogenic technique [22–25], which can significantly reduce the BBR shift and its variations.



An alternative approach was proposed in reference [26] for atoms or ions that have two different clock transitions with frequencies  $\nu_1$  and  $\nu_2$  (e.g. in the ion  $^{171}\text{Yb}^+$ ), that allows the formation of a ‘synthetic’ reference frequency  $\nu_{\text{syn}} = (\nu_1 - \varepsilon\nu_2)/(1 - \varepsilon)$ , which is absent in the spectrum of the involved atoms or ions. In this case, the calibration coefficient  $\varepsilon$  is defined as the ratio of static polarizabilities for transitions with frequencies  $\nu_1$  and  $\nu_2$ , which leads to suppression of the dominating temperature BBR shift term  $\propto T^4$  for the synthetic frequency  $\nu_{\text{syn}}$ . The main advantage of this method is the absence of cryogenic technique and rather moderate requirements for temperature control of the experimental setup, which is very attractive for mobile devices. Apart from  $\text{Yb}^+$ , dual frequency synthesis was also proposed for other systems [27, 28]. In addition, the concept of synthetic frequency was later adapted to suppress various systematic shifts [29–32]. The basic disadvantage of the method [26] is the residual BBR shift due to the higher temperature contributions ( $\propto T^6, T^8, \dots$ ) caused by the dynamic polarizability of atoms (ions). The presence of this residual shift and its variations significantly limits the capabilities of the synthetic frequency method to be in the trend of achieving long-term atomic clock instability at the level of  $10^{-18}$ – $10^{-19}$ .

In this paper, we develop a novel synthetic frequency protocol that allows suppressing the BBR shift variations below  $10^{-19}$  at a given operating temperature  $T_0$  even for unstabilized temperature conditions. This approach actively uses the contribution of the dynamic polarizability of atoms, which leads to the appearance of higher temperature terms ( $\propto T^6, T^8, \dots$ ) in the BBR shift. It is the presence of these terms ( $T^6$ , first of all) that makes it possible to choose the calibration coefficient  $\varepsilon_{T_0}$  so that the average operating temperature  $T_0$  corresponds to a local extremum in the temperature dependence of the BBR shift for the synthetic frequency  $\nu_{\text{syn}} = (\nu_1 - \varepsilon_{T_0}\nu_2)/(1 - \varepsilon_{T_0})$ . In this case, variations of BBR shift become so small that they do not exceed the fractional value of  $10^{-19}$  even at a very low level of thermal control of the experimental setup. In particular, our numerical estimates show that this can be achieved with an atomic clock using  $\text{Yb}^+$  ions for temperature variations in the range  $300 \pm 7$  K.

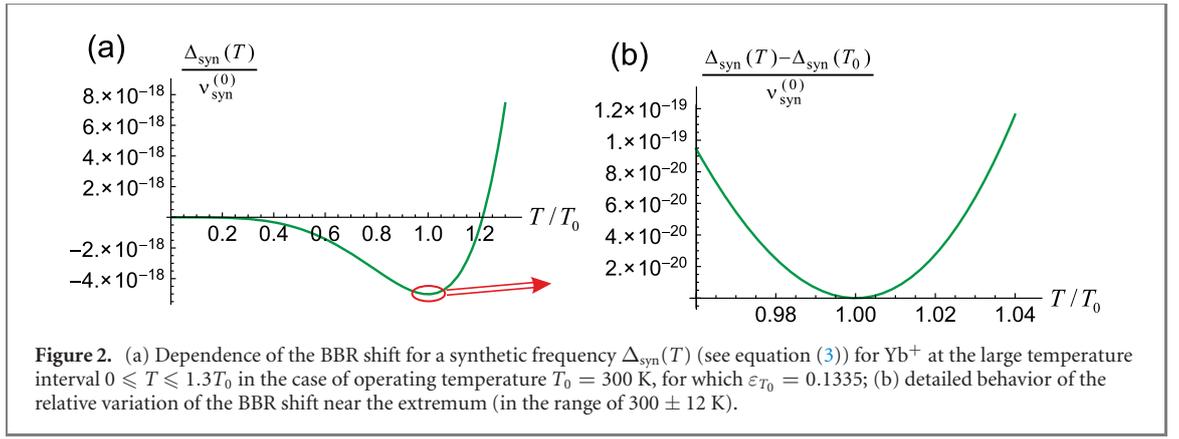
We consider an atom (or ion) in which there are two clock transitions with frequencies  $\nu_1$  and  $\nu_2$  (see, for example, figure 1 for the ion  $\text{Yb}^+$ ). Using formulas from reference [14], it can be shown that BBR shift of atomic energy level is an even function of the parameter  $T$  (from a mathematical viewpoint). In result, the general expression for the frequency of the  $j$ th optical clock transition  $\nu_j(T)$ , which, taking into account the BBR shift  $\Delta_j(T)$ , can be represented as the following series in even powers of the temperature  $T$ :

$$\begin{aligned} \nu_j(T) &= \nu_j^{(0)} + \Delta_j(T), \\ \Delta_j(T) &= a_j \left(\frac{T}{T_0}\right)^4 + b_j \left(\frac{T}{T_0}\right)^6 + c_j \left(\frac{T}{T_0}\right)^8 + \dots, \end{aligned} \quad (1)$$

where  $j = 1, 2$ ;  $\nu_j^{(0)}$  is the unperturbed frequency of the  $j$ th transition; the set of coefficients  $\{a_j, b_j, c_j, \dots\}$  is determined by the structure of the energy levels of the selected atom (ion) and by the operating temperature of the setup  $T_0$ .

Following the synthetic frequency protocol, we consider the superposition

$$\nu_{\text{syn}}(T) = \frac{\nu_1(T) - \varepsilon\nu_2(T)}{1 - \varepsilon}, \quad (2)$$



as the synthetic frequency  $\nu_{\text{syn}}$ , where  $\varepsilon$  is some calibration coefficient. According to equations (1) and (2), the BBR shift of the synthetic frequency is

$$\begin{aligned} \Delta_{\text{syn}}(T) &\equiv \nu_{\text{syn}}(T) - \nu_{\text{syn}}^{(0)} = \frac{\Delta_1(T) - \varepsilon \Delta_2(T)}{1 - \varepsilon} \\ &= \frac{a_1 - \varepsilon a_2}{1 - \varepsilon} \left(\frac{T}{T_0}\right)^4 + \frac{b_1 - \varepsilon b_2}{1 - \varepsilon} \left(\frac{T}{T_0}\right)^6 + \dots, \end{aligned} \quad (3)$$

where  $\nu_{\text{syn}}^{(0)}$  is the unperturbed synthetic frequency

$$\nu_{\text{syn}}^{(0)} = \frac{\nu_1^{(0)} - \varepsilon \nu_2^{(0)}}{1 - \varepsilon}. \quad (4)$$

In reference [26] the calibration coefficient was defined as  $\varepsilon = a_1/a_2$ , which cancels only the main contribution of equation (3) ( $\propto T^4$ ), leaving the higher contributions:

$$\Delta_{\text{syn}}(T)|_{\varepsilon=\frac{a_1}{a_2}} = \frac{b_1 - \varepsilon b_2}{1 - \varepsilon} \frac{T^6}{T_0^6} + \frac{c_1 - \varepsilon c_2}{1 - \varepsilon} \frac{T^8}{T_0^8} + \dots$$

Although this approach suppresses the BBR shift, nevertheless, it is not sufficient from the viewpoint of the current trend toward achieving long-term instability up to the level of  $10^{-19}$ .

Therefore, in this paper, we propose a new strategy for determining the calibration coefficient  $\varepsilon = \varepsilon_{T_0}$ , which depends on the expected operating temperature  $T_0$ . The strength of this approach is that the operating temperature  $T_0$ , arbitrarily chosen as a set point, corresponds to the local extremum of the temperature dependence  $\Delta_{\text{syn}}(T)$  (e.g. see figure 2(a)), i.e.

$$\partial_T \Delta_{\text{syn}}(T)|_{T=T_0} = 0, \quad (5)$$

where  $\partial_T$  denotes the differential operators  $\partial_T = \partial/\partial T$ . Based on equation (3), the condition (5) is achieved for

$$\varepsilon_{T_0} = \frac{\partial_T \Delta_1(T)|_{T=T_0}}{\partial_T \Delta_2(T)|_{T=T_0}} = \frac{4a_1 + 6b_1 + 8c_1 + \dots}{4a_2 + 6b_2 + 8c_2 + \dots}. \quad (6)$$

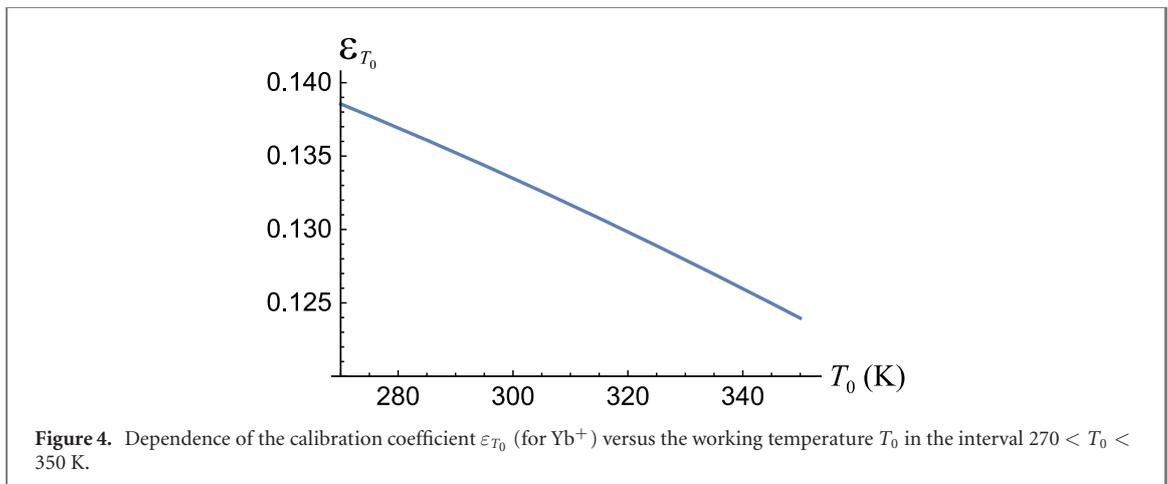
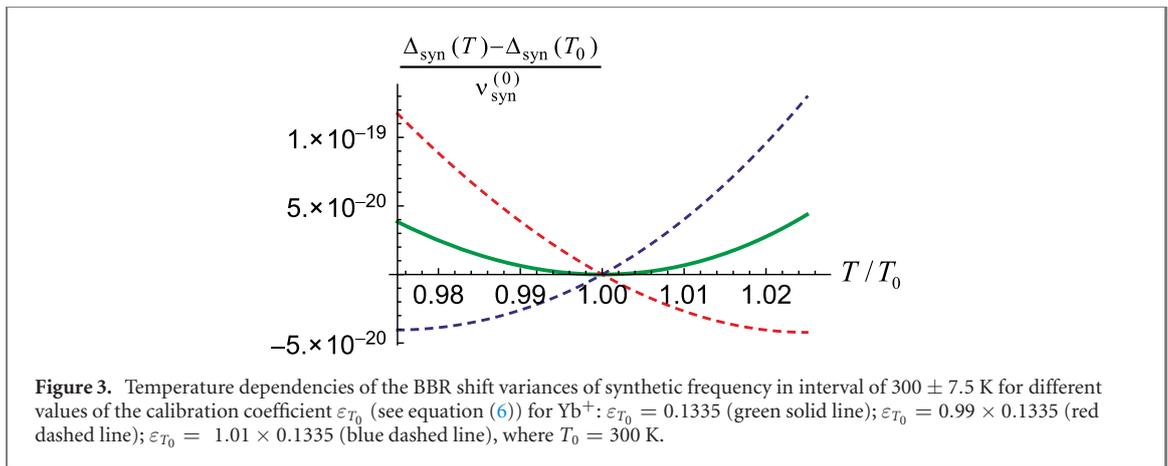
In this case, even relatively large temperature variations will result in only minor variations of the synthetic frequency  $\nu_{\text{syn}}(T)$ .

As an example, consider the ion  $^{171}\text{Yb}^+$ , where we use two narrow-linewidth electric transitions from the ground state in the visible spectral range which can be used as reference transitions of an optical frequency standard (see figure 1): the octupole transition near 467 nm wavelength ( $^2S_{1/2} \rightarrow ^2F_{7/2}$ ,  $F = 0 \rightarrow 3$ ), and the quadrupole transition near 435.5 nm wavelength ( $^2S_{1/2} \rightarrow ^2D_{3/2}$ ,  $F = 0 \rightarrow 2$ ). Alternatively, the ion provides an additional quadrupole transition near 411 nm which could be used in even isotopes [33]. In the case of operating temperature  $T_0 = 300$  K, we will use the BBR shift estimates for the octupole transition based on measurements in reference [34]:

$$a_1 = -0.04537 \text{ Hz}, \quad b_1 = 6.8 \times 10^{-5} \text{ Hz}, \quad (7)$$

and also our calculations for the quadrupole transition based on data from reference [35]:

$$a_2 = -0.2826 \text{ Hz}, \quad b_2 = -0.0322 \text{ Hz}, \quad c_2 = -0.00321 \text{ Hz}. \quad (8)$$

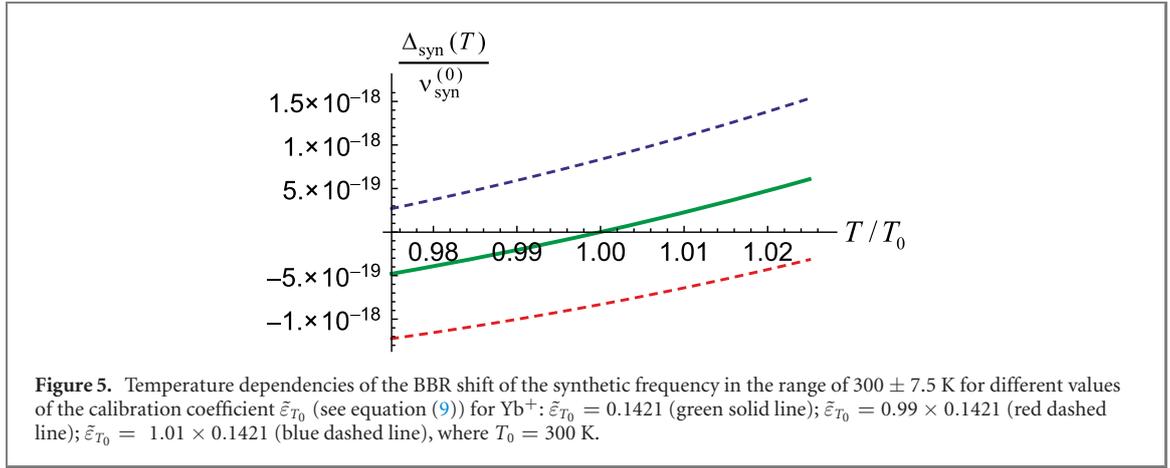


In this case, using equation (6), we find  $\varepsilon_{T_0} = 0.1335$ . For comparison, if we want to cancel in equation (3) only the main contribution ( $\propto T^4$ ) we need to use another calibration coefficient  $\varepsilon = a_1/a_2 = 0.1606$ , which does not depend on the operating temperature  $T_0$  and significantly differs from  $\varepsilon_{T_0}$ .

Figure 2(a) shows the behavior of the BBR shift (see equation (3)) for a synthetic frequency over a large temperature range ( $0 \leq T \leq 390$  K), where an extremum at the operating temperature  $T_0$  is clearly visible. Figure 2(b) contains the detailed behavior of this shift near the extremum: although at the point  $T_0$  the BBR shift itself has a relative magnitude  $\Delta_{\text{syn}}(T_0)/\nu_{\text{syn}}^{(0)} = -5 \times 10^{-18}$  (see figure 2(a)), its variations do not exceed  $10^{-19}$  over a relatively large temperature interval of  $300 \pm 12$  K.

However, atomic structure calculations and the measurements on which we both base the determination of the expansion coefficients of equations (7) and (8) are of limited accuracy. Figure 3 demonstrates how this can affect the suppression of variations in the BBR shift, if the accuracy of atomic calculations for  $\varepsilon_{T_0}$  is 1% for the  $\text{Yb}^+$  ion. In this case, BBR shift variations will not exceed the relative value of  $10^{-19}$  in the temperature range  $300 \pm 7$  K, which still corresponds to a very low level of temperature control of the experimental setup. For comparison, if we consider the bare octupole transition  $^2S_{1/2} \rightarrow ^2F_{7/2} (F = 0 \rightarrow 3)$  near 467 nm wavelength, the relative BBR shift is  $-7.1 \times 10^{-17}$  at the operating temperature  $T_0 = 300$  K with a variation of about  $\pm 8 \times 10^{-18}$  within  $\delta T = \pm 7$  K. In the case of the bare quadrupole transition  $^2S_{1/2} \rightarrow ^2D_{3/2} (F = 0 \rightarrow 2)$  near 435.5 nm wavelength, the relative BBR shift is  $-4.1 \times 10^{-16}$  for  $T_0 = 300$  K, and its variation is approximately  $\pm 4 \times 10^{-17}$  within  $\delta T = \pm 7$  K. These comparisons show that our method radically reduces the effect of temperature variations on the long-term stability in an atomic clock by at least two orders of magnitude. This will make it possible to abandon the use of cryogenic technology even in devices with a very low level of temperature control, i.e. outside of laboratory conditions.

Obviously, for any other operating temperature  $T_0$ , all the above calculations can be easily redone (see figure 4). Note also that our semi-phenomenological calculations for  $\text{Yb}^+$ , based on the estimates in equations (7) and (8), do not claim high accuracy (e.g. see reference [36]) and are used primarily to demonstrate the high potential of our method for ultra-precise atomic clocks. Moreover, theoretical



calculations can be combined with additional experiments that make it possible to refine the value of  $\varepsilon_{T_0}$  in each specific case.

We can consider another option for determining the calibration coefficient  $\varepsilon = \tilde{\varepsilon}_{T_0}$  in such a way as to zero the BBR shift for the synthetic frequency equation (3) at the operating temperature  $T_0$ :

$$\Delta_{\text{syn}}(T_0)|_{\varepsilon=\tilde{\varepsilon}_{T_0}} = 0 \Rightarrow \tilde{\varepsilon}_{T_0} = \frac{\Delta_1(T_0)}{\Delta_2(T_0)} = \frac{a_1 + b_1 + c_1 + \dots}{a_2 + b_2 + c_2 + \dots}. \quad (9)$$

For example, for  $\text{Yb}^+$  we find  $\tilde{\varepsilon}_{T_0} = 0.1421$  at the operating temperature  $T_0 = 300$  K. Figure 5 shows (see solid green line) the temperature dependence of the BBR shift in this case. Also shown is the effect of inaccuracy (at the 1% level) in determining the calibration coefficient  $\varepsilon = \tilde{\varepsilon}_{T_0}$  (see blue and red dashed lines in figure 5). In general, comparing figures 3 and 5, we can conclude that, from the viewpoint of minimizing sensitivity to temperature variations, the calibration coefficient according to equation (6) is more preferable than equation (9), at least for  $\text{Yb}^+$ .

Generalizing the presented options, we can consider a combined atomic clock using three different clock transitions with frequencies  $\nu_1$ ,  $\nu_2$  and  $\nu_3$ . In this case, the synthetic frequency is defined as the superposition

$$\nu_{\text{syn}}(T) = \frac{\nu_1(T) - \varepsilon_2 \nu_2(T) - \varepsilon_3 \nu_3(T)}{1 - \varepsilon_2 - \varepsilon_3} \quad (10)$$

with two calibration coefficients  $\varepsilon_2$  and  $\varepsilon_3$ . Here, the strategy is the simultaneous fulfillment of two conditions

$$\Delta_{\text{syn}}(T_0) = 0, \quad \partial_T \Delta_{\text{syn}}(T)|_{T=T_0} = 0, \quad (11)$$

i.e. the absence of the BBR shift (at the point  $T_0$ ) and its very weak sensitivity to temperature variations near the operating temperature  $T_0$ . These conditions correspond to the calibration coefficients  $\varepsilon_2$  and  $\varepsilon_3$  to satisfy the equations

$$\begin{aligned} \varepsilon_2 \Delta_2(T_0) + \varepsilon_3 \Delta_3(T_0) &= \Delta_1(T_0), \\ \varepsilon_2 \partial_T \Delta_2(T)|_{T=T_0} + \varepsilon_3 \partial_T \Delta_3(T)|_{T=T_0} &= \partial_T \Delta_1(T)|_{T=T_0}, \end{aligned} \quad (12)$$

where a solution can be always obtained at an arbitrary operating point  $T_0$ . Such a condition can be found in composite clock systems or in single species clocks such as  $\text{Yb}^+$  and  $\text{Lu}^+$  [37], where a single ion offers three different clock transitions.

Note that the synthetic frequency  $\nu_{\text{syn}}$  can be realized either virtually or as a real frequency component of a femtosecond comb generator [26]. In the case of  $\text{Yb}^+$ , we find the synthetic frequency  $\nu_{\text{syn}} = 635$  THz (for  $\varepsilon_{T_0} = 0.1335$ ), corresponding to a wavelength  $\lambda_{\text{syn}} = 472$  nm. This frequency lies sufficiently close to the initial reference transitions at 435.5 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. With this, a synthetic frequency that is robust against temperature fluctuations is realized as an optical reference. On the one hand, this makes it possible to experimentally compare the combined clocks located in different laboratories. On the other hand, this allows us to potentially consider a synthetic frequency for a new definition of the second. To do this, the metrological community must select a specific type of combined clock (i.e. a pair of initial clock transitions  $\nu_1$  and  $\nu_2$ ), then to fix the operating temperature  $T_0$  and the corresponding calibration

coefficient  $\varepsilon_{T_0}$ . The residual BBR shift  $\Delta_{\text{syn}}(T_0)$  (see in figure 2(a)) should also be taken into account using theoretical calculations and/or experimental measurements.

In conclusion, we have developed a synthetic frequency method to drastically suppress BBR shift variations in an atomic clock under normal environmental conditions at an arbitrary operating point  $T_0$ . In this case, the efficiency of the method improves by one to two orders of magnitude compared to previously published work. Our approach is an alternative to the use of cryogenic technologies to achieve long-term instability of  $10^{-18}$ – $10^{-19}$  in ultra-precision atomic clocks. This can be especially attractive for portable devices due to the lack of strict requirements for temperature stabilization. On the other side, if lower accuracy but improved long-term stability is required, such as in improved frequency standards for industrial applications, the two electric quadrupole transitions near 435.5 nm and 411 nm in  $\text{Yb}^+$  could be used to engineer a low drift and temperature independent frequency standard, outdating the currently used hydrogen maser.

In general, the method can be easily adapted to other clock transitions in atoms (ions), such as the near-411 nm quadrupole transition in combination with the octupole transition in the even isotope  $^{172}\text{Yb}^+$ , where precision spectroscopy has been recently reported [33]. Also, multi-species optical clocks could benefit from the method. There, the synthetic frequency can consist of a superposition of one clock transition within each of the species: neutral atom pair Sr/Yb in the same vacuum chamber using  $^1\text{S}_0 \rightarrow ^3\text{P}_0$  clock transitions [38]; or ion pairs  $\text{In}^+/\text{Yb}^+$ ,  $\text{Sr}^+/\text{Yb}^+$ ,  $\text{Yb}^+/\text{Ca}^+$  and  $\text{Hg}^+/\text{Sr}^+$  [19, 28, 39]. In the second case, both ions benefit from the sympathetic cooling properties to reach lower ion temperatures. The synthetic frequency is then derived from long-lived clock states in each of the species, that could even be simultaneously interrogated. In addition, our method can serve as a stimulus for the development of numerical methods for atomic calculations of the BBR shift (including dynamic contributions) with an accuracy of 1% for the correct determination of the calibration coefficient.

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## Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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