

Cesium atomic Doppler broadening thermometry for room temperature measurement

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Atomic Doppler broadening thermometry (DBT) is potentially an accurate and practical approach for thermodynamic temperature measurement. However, previous reported atomic DBT had a long acquisition time and had only been proved at the triple point of water, 0°C, for the purpose of determination of the Boltzmann constant. This research implemented the cesium atomic DBT for fast room temperature measurement. The Cs¹³³ D1 (6S^{1/2} → 6p^{1/2} transition) line was measured by direct laser absorption spectroscopy, and the quantity of thermal-induced linewidth broadening was precisely retrieved by the Voigt profile fitting algorithm. The preliminary results showed the proposed approach had a 4 min single-scan acquisition time and 0.2% reproducibility. It is expected that the atomic DBT could be used as an accurate, chip-scale, and calibration-free temperature sensor and standard.

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With the determination of the Boltzmann constant, k_B , the 26th General Conference on Weights and Measures passed the No. 1 resolution, which redefined the International System of Units (SI) of thermodynamic temperature, kelvin (K), to this fundamental physical constant^[1,2]. The redefinition of the unit of temperature will bring merging and a transition from the current international temperature scale to the thermodynamic temperature scale in the long term. In the draft of ‘Mise en Pratique of the Realization of the Kelvin (MEP-K) 2019’^[3], the Consultative Committee for Thermometry defines four primary thermometry approaches, which are acoustic gas thermometry^[4,5], dielectric-constant gas thermometry^[6], Johnson noise thermometry^[7], and absolute radiative thermometry^[8] in a metrological narrow sense. However, the four metrological primary-purposed approaches encounter practical issues in terms of having complicated systems and long acquisition times. Therefore, practical and accurate methods for thermodynamic temperature measurement need to be developed.

Doppler broadening thermometry (DBT) measures the thermodynamic temperature from the quantity of line broadening of an atomic or molecular absorption spectrum. The broadening is induced by statistically distributed velocities of the particles under their thermal equilibrium states. It is a primary thermometry approach since it obtains the thermodynamic temperature instead of the international scaled temperature. Compared to other novel thermodynamic temperature measurement approaches^[9,10], it has less perturbation sources and a relatively clear uncertainty budget. In 2002, Borde first proposed that the measurement of Doppler broadening could

be a practical and accurate approach to determine the Boltzmann constant^[11]. In 2007, Daussy *et al.* implemented the first Boltzmann-constant measurement by DBT with direct laser absorption spectroscopy, where the ¹⁴NH₃ ν_2 band Q(6,3) vibration-rotation line was observed and fitted by the Gaussian profile. The relative uncertainty of the measurement was 190 ppm (parts per million)^[12,13]. After that, an improved system was established in 2011. The soft collision effect was accounted for, and the Galatry profile was used to fit the spectra. Thereafter, a 6.4 ppm statistical uncertainty result was obtained^[14]. In 2013, Daussy *et al.* further improved the budget of type B uncertainty by introducing the speed-dependent Voigt profile (SDVP) and obtained a 2.3 ppm type B result^[15]. In 2008, Gianfrani *et al.* precisely recorded the line shape of the R(12) $\nu_1 + 2\nu_2^0 + \nu_3$ transition in CO₂ gas. The Boltzmann constant was determined using the Voigt line model, and the corresponding relative deviation was 1.6×10^{-4} ^[16]. In 2011, Gianfrani *et al.* observed the speed-dependent effect on the profile of $4_{4,1} \rightarrow 4_{4,0}$ of the H₂¹⁸O $\nu_1 + \nu_3$ band. They used the speed-dependent Galatry profile to improve the measurement. It was the first time that DBT was proposed for temperature measurement^[17]. In 2013, Gianfrani *et al.* used the partially correlated speed-dependent hard-collision profile (PCSDHCP) to give a relative standard uncertainty result of 24 ppm^[18]. In 2018, they conducted a thermodynamic temperature measurement on the Pe(12) line of CO₂ using cavity ring-down spectroscopy (CRDS) rather than direct absorption spectroscopy and obtained a result on the order of 10 ppm^[19]. Since 2010, Hu *et al.* made intensive researches on k_B measurement by CRDS. They used the R(9) line in the $\nu_1 + 3\nu_3$

band of $^{12}\text{C}_2\text{H}_2$ and got a statistical uncertainty result of 12 ppm^[20–22]. In addition to all the above-mentioned researches on molecular DBT, Stace and Luiten determined k_B with direct laser absorption spectroscopy on dilute alkali atomic vapor in 2010^[23]. At the triple point of water and low probe laser power, saturation broadening was depicted and mitigated by introducing a nonlinear Voigt factor to the Lambert–Beer law. An uncertainty of 71 ppm for the determination of k_B was reported in 2015^[24,25].

In recent years, the leading national metrology institute is developing chip-scale measurement standards based upon the quantum theories and microelectromechanical system (MEMS) technologies^[26]. It is expected that, by taking advantage of the interaction between light and matter or structures, the conventional electric measurement would be substituted by photonic approaches, by which a fast, on-site, calibration-free, and multi-physical measurement platform can be implemented. Atomic DBT is a promising approach for the compact and accurate solution for thermodynamic temperature measurement, because the atom compared with the molecule has a large absorption coefficient and, therefore, a high signal-to-noise ratio in detection. In previous reports, k_B measurements by means of alkali atomic DBT had only been verified at the triple point of water, but not for the other temperature points^[24,25]. Besides, for pursuing the high accuracy, it took a long time for noise mitigation, which is not practical in a real out-of-lab scenario^[27,28]. This research proved that the cesium atomic DBT could measure the thermodynamic temperature in the room temperature range with a short acquisition time. For a single scan, the acquisition time was less than 4 min, and measurement reproducibility was 0.2%.

Direct laser absorption spectroscopy turns the probe laser in a narrow spectral band with a single through or multiple reflections in a sensing unit to acquire the transition spectrum of atomic or molecular gas. The attenuation of the beam intensity is characterized by the Lambert–Beer law. Under thermal equilibrium conditions, according to the Doppler effect, the relative velocity of a gas particle brings a spectral red or blue shift. It is statistically shown as spectrum broadening, the Doppler broadening. The full width at half-maximum of the absorption line and the thermodynamic temperature has the relationship

$$\Gamma_D = \frac{\nu_0}{c} \sqrt{8 \ln 2 \frac{k_B T}{m}}, \quad (1)$$

where ν_0 is the central frequency of the absorption line, c is the speed of light in the vacuum, k_B is the Boltzmann constant, T is the thermodynamic temperature, and m is the mass of the particle. The atomic absorption spectrum was affected by Doppler broadening, pressure broadening, self-collisional broadening, etc. For a dilute atomic vapor in an ultra-high vacuum cell, the absorption line can be depicted using the Voigt line shape, which is a convolution of the

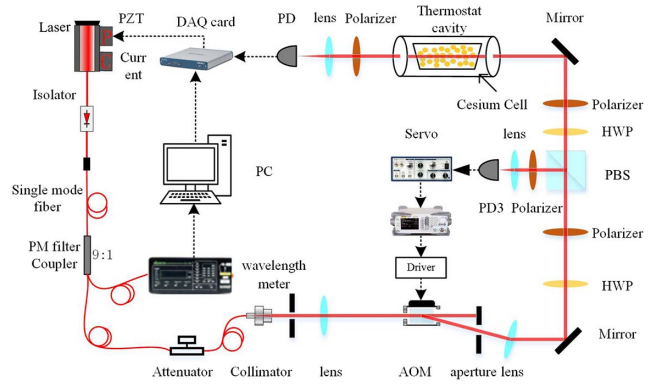


Fig. 1. Cs^{133} DBT experimental setup.

Gaussian and Lorentzian profiles. Atomic DBT deconvolutes the two line functions and extracts the Doppler components to obtain the thermodynamic temperature by Eq. (1).

The system setup is shown in Fig. 1. An external cavity diode laser (Newfocus TLB6718) was used as a probe laser. The output beam was connected to a 90:10 polarization-maintaining fiber splitter. The minor component was guided to a high-precision wavelength meter (Topical WSU-10IR1) for frequency monitoring and control. The probe beam was collimated and incident to an amplitude modulated acousto-optic modulator (AAOptoelectronic, MT250-B100A0, 5-8001001), which was driven by a radio frequency signal generator (Rigol DSG815). The intensity of the first diffracted order was actively stabilized through a proportional-integral servo controller (Newfocus LB1005). The frequency stability and amplitude stability of the probe beam were 2×10^{-9} at 100 s and 6×10^{-4} , respectively. A high vacuum cesium vapor cell (Triad Technologies TT-CS-25 \times 75-Q-BA) with 10^{-7} Torr (1 Torr = 133.322 Pa) pressure was placed in a thermostat. Two calibrated PT100 platinum resistance thermometers were placed at both ends. One was a control sensor, and the other was a measurement sensor, which provided the reference value of the verification temperature. The thermostat was electrically heated and controlled by a precision temperature controller (Meerstetter, TEC-1091), and the reference temperatures were measured by a precision thermometer (Anton Parr, MKT50). A 6 GHz frequency detuning with a 20 MHz scan interval was controlled by self-developed software with a high-speed data acquisition card (NI USB-6361). To the cesium D1 line, the span was wide enough to capture the expected features and noise characteristics of the absorption spectrum. The total acquisition time is less than 4 min, and it could be decreased by taking a large scan interval.

With a probe power of 110 μW , the absorptions of the F4-3 and F4-4 lines are shown in Fig. 2.

The Voigt profile was fitted with the value of the natural linewidth of the $6\text{S}^{1/2} \rightarrow 6\text{p}^{1/2}$ transition^[29], where the Gaussian and Lorentzian components were shared between the above two absorption features. The influence

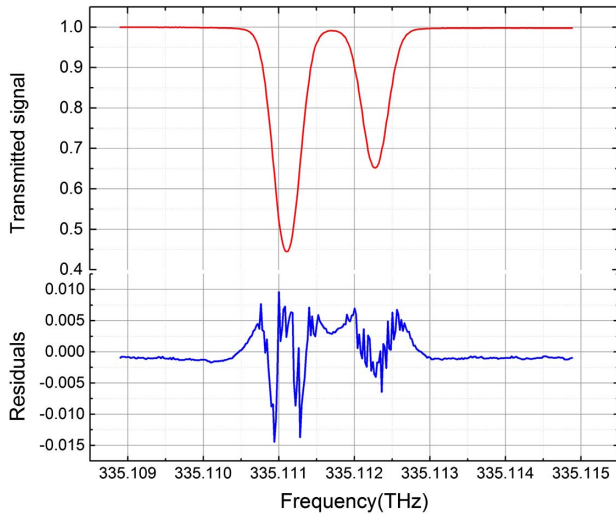


Fig. 2. Cs D1 absorption line and fitted residuals near the probe power of $110 \mu\text{W}$ at a verification temperature point of 301.526 K .

of the self-collisional broadening could be neglected, since the temperature of the vapor cell determined that component is 460 Hz , which is a quite small amount compared to the reference value of the D1 natural linewidth. Further, as the vapor cell did not have any buffer gas, there is no pressure broadening. A 200 kHz laser-induced broadening on the Lorentzian component was accounted for in the fitting calculation. With the above considerations, the extracted width of the Doppler broadening was 361.48063 MHz , and its maximum fitting residual was 1.5% , as shown in Fig. 2. According to Eq. (1), the measured thermodynamic temperature was 301.46 K . For a single scan, this was the best result among a set of sixty experiments. Compared to the measured reference temperature of 301.526 K , it had a relative error of -0.02% .

In order to verify the saturation broadening effect, a bunch of measurements was conducted within a probe power range between 5 and $170 \mu\text{W}$. Figure 3 plots the relative errors of the measurement results as a function

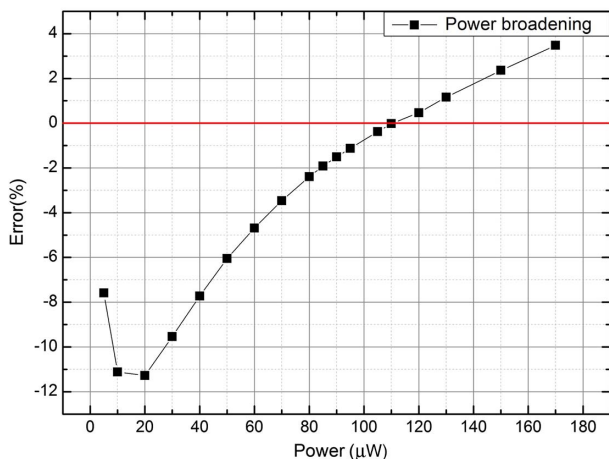


Fig. 3. Error of measurement as a function of probe power.

of the powers. At each point, 25 measurements were conducted, and the results were averaged. The figure showed a quasi-linear relationship between the measurement errors and probe power within $50\text{--}130 \mu\text{W}$.

A set of sixty consecutive experiments was conducted at the $110 \mu\text{W}$ probe power. The average in terms of relative error and the relative standard deviation are shown in Fig. 4. It is shown that a short-term type A uncertainty of 0.15% was preliminarily obtained.

With regard to the reproducibility, we conducted two sets of independent experiments under the same condition and examined the results of the linear region, $50\text{--}130 \mu\text{W}$, in terms of the error–power relationship. As shown in Fig. 5, the maximum difference of the two measurements within this region was less than 0.2% .

The above preliminary results proved that the cesium atomic DBT could measure the thermodynamic

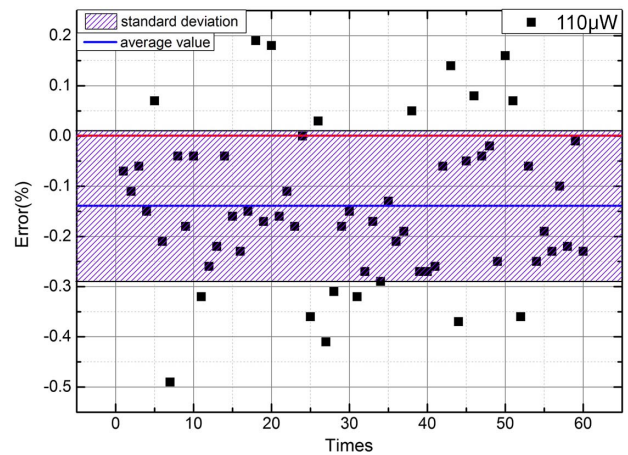


Fig. 4. Results of sixty measurements with the probe power of $110 \mu\text{W}$ at the reference temperature of 301.526 K . The blue straight line shows the relative error of the averaged results. The shade depicts the relative standard deviation of the measurements, and the red line shows the reference value.

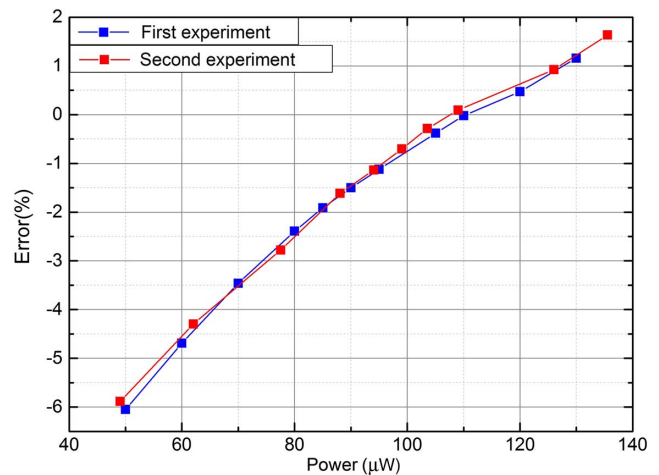


Fig. 5. Verification of reproducibility by comparing the results of two independent measurements within the linear region.

temperature at room temperature. With the verification system, the broadening of the spectrum caused by self-collision, atom–molecule collision, and external magnetic fields can be neglected when compared to the megahertz (MHz) scale Doppler and natural linewidth. Further, the measured spectra were obtained with a single scan rather than multiple averages, and the curve fitting did not consider nonlinear power broadening. Therefore, it is plausible that the maximum amplitude of the fitted residual reached 1.5% level. The probe power where the measurement obtained the best accuracy was experimentally determined as 110 μW . By introducing a quadra-Voigt term to the Lambert–Beer law or other advanced line profiles, the slant of the error–power curve within the linear region can be reduced, and measurement error could be mitigated to a relatively low value, less than 0.5%, within a large power range. Therefore, high-accuracy and calibration-free sensing can be implemented. It is worth noting that the standard deviation of 0.15% of the results of the short-term repeated experiments and the reproducibility of 0.2% in the linear region could match with each other. The overall performance can be improved by means of further stabilizing the probe frequency and power, and the uncertainty budget can be obtained by taking in the above-mentioned minor components.

In conclusion, we proved that atomic DBT based on direct laser absorption spectroscopy could be used to measure the thermodynamic temperature in the room temperature range with a short acquisition time. The transition absorption spectra of the cesium D1 ($6S^{1/2} \rightarrow 6P^{1/2}$) line were measured. The Doppler width and corresponding thermodynamic temperature were extracted by the Voigt profile fitting algorithm. Without power broadening correction, the best relative error of a single scan of the measurement was -0.02% , and the reproducibility was better than 0.2%. In the future, for implementing a calibration-free thermodynamic temperature measurement, power broadening must be taken into account, and the advanced line profile could be used to mitigate the error within a large probe power range. We expect that with MEMS technologies, in terms of fabricating millimeter-sized vapor cells, atomic DBT can implement an accurate, chip-scale, and calibration-free thermodynamic temperature sensor and standard.

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