Progress report for the European Metrology Programme for Innovation and Research (EMPIR) project, Realising the Redefined Kelvin, "Real-K"

Prepared: Graham Machin, NPL

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Introduction

This is the second progress report to the CCT of the EMPIR Realising the Redefined Kelvin project (Real-K).

The aim of Real-K is to take the kelvin redefinition, and its associated *mise-en-pratique* and begin to turn it into a reality by:

a) developing primary thermometry techniques at high and low temperatures which can be used to realise and disseminate the redefined kelvin b) perform research to ensure the on-going fitness of the ITS-90, and c) undertake longer-term research for gas based primary thermometry to be the basis for temperature traceability throughout the entire range.

The project began on 1 September 2019 and is due to run until March 2023. The project website address is https://real-k.aalto.fi. The project partners are: NPL, (United Kingdom, coordinator), Aalto University (Finland), CEM, (Spain), CMI, (Czech Republic), CNAM, (France), Fondazione Bruno Keller (FBK, Italy), Helmut Schmidt Universitat (HSU, Germany), INRiM, (Italy), INTiBS, (Poland), IPQ, (Portugal), LNE, (France), NIM, (China), PTB, (Germany), SMU, (Slovak Republic), TIPC-CAS, (China), TUBITAK, (Turkey), UL, (Slovenia), Uniwersytet Warszawski (UW, Poland), VNIIOFI, (Russia). and VTT, (Finland). Unfortunately, VSL, (The Netherlands) had to leave the project because of technical capability issues.

Objectives

The objectives of the project are:

1) To establish and demonstrate traceability directly to the redefined kelvin, by indirect primary radiometry, from ~1300 K to ~3000 K. Low uncertainty thermodynamic temperatures of four new HTFPs will be determined. Then, through the *MeP*-K-19, HTFPs will be used to realise and disseminate thermodynamic temperature with uncertainties competitive with the ITS-90 ($U<0.05\%$).

2) To develop practical primary thermometry for realisation and dissemination of thermodynamic temperature below 25 K and demonstrate that primary thermometry can be used: to replace the currently complex ITS-90 scale realisation arrangement below 25 K and to ensure a smooth transition to the PLTS-2000 range below 1 K (U = 0.2 mK at 25 K and <1% at 1 K).

3) To extend the life of the current defined scale (ITS-90) giving users continued access to low uncertainty realisations of the scale whilst allowing time for primary thermometry methods to mature. Scale non-uniqueness will be investigated, with the objective of reducing its uncertainty by 30 %, and a suitable fixed-point replacement for the mercury triple point identified, constructed and tested. Integration of the new fixed point whilst retaining the ITS-90 framework will be investigated.

4) To reduce the uncertainty in a number of different primary thermometry methods, approved for use in the *MeP*-K-19, and so begin to facilitate their applicability for temperature realisation

and dissemination into the temperature region 25 K and above. Here the uncertainties of the calculated thermophysical properties of gases (e.g. He, Ne, Ar) used as thermometric fluids in primary thermometers will be reduced, selected values will be confirmed by measurements.

Results to date

Key results are given below after approximately two years of research, however approximately 6 months were lost due to the Covid-19 pandemic.

Realisation and dissemination of the redefined kelvin >1300 K

The *current state of the art* for temperature realisation and dissemination >1300 K is through the ITS-90. The state of the art will be advanced by assigning definitive thermodynamic temperatures to the High Temperature Fixed Points (HTFPs): Fe-C (1426 K), Pd-C (1765 K), Ru-C (2226 K) and WC-C (3020 K), and by demonstrating the first practical outworking of the *MeP*-K-19 by indirect primary radiometry (>1300 K).

The following high temperature fixed points have been constructed in line with the construction protocol; 5 WC-C cells, 4 Ru-C cells, 6 Ru-C cells, 5 Pd-C cells and 4 Fe-C cells. This is in excess of the minimum required by the project, but it allows for some redundancy in case of cell breakage.

A protocol has been agreed for the measurements required to characterise the thermal effects on the cell reproducibility and relevant measurement apparatus has been prepared to undertake these evaluations. The partners, along with a company in Spain have been identified to perform dissemination trials later in the project.

Presently, two batches of cells at the Fe-C, Pd-C, Ru-C and WC-C points were formed with the best cells produced identified after a thorough characterisation and comparison of their performance. These cells are being circulated in 8 NMIs to achieve a low-uncertainty determination of their thermodynamic phase-change temperatures.

Realisation and dissemination of the redefined kelvin <25 K

The *current state of the art* for the temperature scale <25 K is complex. Traceable temperatures are referenced to ITS-90 or PLTS-2000. Scale realisation requires different sophisticated, time-consuming, experimental methods, which are only available in a few National Metrology Institutes (NMIs). The state of the art will be advanced through developing primary thermometers to cover the entire range from 1 K to 25 K for direct realisation and dissemination of temperature (target uncertainty <1 %).

For the Johnson Noise Thermometer, the primary Magnetic Field Fluctuation Thermometer (pMFFT) was redesigned and has now been constructed. The main uncertainty component of the noise temperature, which arises from determination of geometrical parameters, should be reduced through this design, hopefully by at least a factor 2. Preliminary measurements using a pMFFT have been performed at the helium lambda point (2.1768 K), the temperature at which normal fluid helium transitions to superfluid helium.

The nanofabrication process for Ge based Coulomb Blockade Thermometers (CBTs) was modified and devices have been constructed with 100 nm Al junctions. These have been tested and shown to successfully operate between 1 K and 25 K. This is a very promising first step to utilising CBT as primary thermometers over the entire temperature range specified in the project.

The pulse-tube cryostat between 4 K and 24 K that was installed in a cryostat belonging to LNE/CNAM for fast-Acoustic Gas Thermometry (fast-AGT) and it is now fully operational. LNE/CNAM and TIPC-CAS have worked together and jointly developed “fast-AGT” measurement techniques at microwave frequencies, which will be used on a single-pressure refractive-index thermometer (SPRIGT) installed at TIPC-CAS.

Extending the life of the International Temperature Scale of 1990

The current state of the art leaves Type 1[1] and 3[2] non-uniqueness among the dominant uncertainties in ITS-90 calibrations. However significant data to support uncertainty contribution assignment is only available in limited sub-ranges of the ITS-90 e.g. 0 ºC to 420 ºC. There are three candidates for replacing the hazardous mercury triple point, namely Xe, CO_2 or SF_6. The state of the art will be advanced through substantially increasing knowledge of Type 1 and 3 non-uniqueness in ITS-90 calibrations (target 30 % uncertainty reduction) and in characterising at least two suitable replacements for the mercury triple point. Great progress has been made on examining resistance ratio data from the Ar to Zn fixed points and this has allowed Type 1 non-uniqueness uncertainties to be quantified in detail resulting in a paper being recently submitted to Metrologia.

Robust evaluation of non-uniqueness began with eight long stem platinum resistance thermometers (LSPRTs) being compared and calibrated. Two of these were transported to join a batch of four others for detailed evaluation at a specialist partner. Unfortunately, this partner had to withdraw from the consortium and the thermometers were transported to a new partner taking on their role. These measurements have subsequently been completed and were transported to the next partner for SPRT non-uniqueness evaluation down to approximately -190 ºC. Due to the pandemic, work on the new gas-controlled heat pipe, for the comparison of high temperature SPRTs, for evaluating the type 3 non-uniqueness in the range between the Al and Ag fixed points, was paused, but is now underway.

With regards to investigating alternatives to the Hg triple point:

For the construction of CO_2 fixed point cells, a cylinder of very high purity (99.9999 %) gas was purchased from Linde. Two cells were manufactured from stainless steel and checked for their durability up to 60 bar. They were filled with high purity CO_2 gas using a Static Gas Mixture Preparation system and initial measurements were performed. The initial results were found to be promising in terms of plateau stability and reproducibility. A preliminary chemical analysis of the CO_2 was performed using Gas Chromatography.

Three sealed cells for the realisation of the SF_6 triple point have been manufactured. A calorimeter dedicated to the measurement of the SF_6 triple point that allows a simultaneous calibration of long-stem and capsule-type SPRTs has been constructed. The cells fit in a different cryostat at the institute allowing a detailed investigation of the effect of the thermal environment on the SF_6 triple point to be performed. This is important as it is thought to be the main source of uncertainty.

An investigation has begun into what impact the removal of the Hg triple point from the ITS-90 will have on its interpolating equations and uncertainties. Currently relevant literature connected with the use of SF_6 and CO_2 as alternative temperature fixed points to the Hg triple point is being examined. This study will include an examination of the effect of not replacing the Hg point and its impact on the realisation of ITS-90.

Facilitating full range primary thermometry

The *current state of the art* in establishing traceable temperatures is through calibration to, e.g. the ITS-90. For primary thermometry to be practical for kelvin dissemination between 25 K and 1300 K, gas-based methods, i.e. AGT, Dielectric Constant Gas Thermometry (DCGT) and RIGT, need to be substantially simplified. The state of the art for gas based primary thermometry will be advanced through reducing the *ab initio* calculation uncertainty of the non-

[1] Type 1 non-uniqueness – arises from the use of different equations in overlapping ranges, using the same thermometer

[2] Type 3 non-uniqueness – arises from the use of different interpolating thermometers of the same kind in the same range

ideality of monatomic gases He, Ar and Ne, with the calculations validated by low-uncertainty measurements over a temperature range of (10 K to 350 K) and pressures (<100 MPa).

Modelling

The second virial coefficient *B(T)* and second acoustic virial coefficient for helium-3 and helium-4 have been computed between 1 K and 400 K using a highly accurate pair interaction potential involving a new representation of the relativistic and quantum-electrodynamics (QED) components. The results of these calculations represent a significant, about six-fold improvement in accuracy compared to the previous *ab initio* calculations. Remarkable agreement between the values and uncertainties of *B(T)* has been obtained by two different partners, using independently written code providing important cross-validation of these results.

The non-additive three-body interaction energy for helium has been represented as a sum of three contributions: the nonrelativistic Born-Oppenheimer (BO) energy, the adiabatic correction due to the coupling of nuclear end electronic motion, and the relativistic correction. The latter two effects have not been previously considered in the literature for this system. The expected error of the three-body potential is less than 1 percent. This activity is expected to be completed in 2021.

The software needed for the evaluation of the third density virial *C(T)* of He, Ne and Ar and the transport properties of Ne and Ar, has been updated and optimised. It is now ready to perform the calculations once the pair and three-body potentials developed within the same task are finalised.

A path-integral Monte Carlo (PIMC) method and state-of-the-art two-body and three-body potentials were used to calculate the fourth virial coefficients *D(T)* of ^{4}He and ^{3}He as functions of temperature from 2.6 K to 2000 K.

Measurement of selected thermophysical properties of gases

Speed of sound acoustic data previously measured using an AGT has yielded values of the second acoustic virial coefficient of argon in the range 120 K to 330 K. The values were in remarkable agreement with *ab-initio* calculations. Work is in progress to estimate the uncertainty of these results.

The density virial coefficients of helium were determined by coupled DCGT and Burnett expansion experiments at 273 K and 296 K including corrections for e.g. effects of pressure deformation of the vessels containing the measuring gas. The relative uncertainty of the second density virial coefficient of helium is ~2.5 % whereas the deviation to the values are <1 % proving the performance of the apparatus. In addition, similar measurements 253 K, 273 K, 296 K and 303 K with argon have also been completed. The uncertainty for the second density virial coefficient was <1 % and agreed well with available literature values with deviations less than the standard uncertainty. Measurements with neon are currently being prepared.

Speed-of-sound measurements in supercritical neon gas were carried out with a double-path-length pulse-echo technique. Measurements were taken, along 12 supercritical isotherms between 200 K and 420 K, every 20 K, at pressures between 20 MPa and 100 MPa. The combined expanded uncertainty of the speed of sound was 0.007 %, including the contributions of the uncertainty of the temperature and pressure measurements.

Preparing for improved primary thermometry

Modification and adaptation for AGT of a cryogenic apparatus (previously used for RIGT) has started with tests of the acoustic transduction systems between 12 K and ambient temperature in an acoustic resonator.

The refractive index of helium using a quasi-spherical copper microwave resonator at five different temperatures between the triple point of hydrogen at 13.8 K and the triple point of Xe at 161.4 K for pressures up to 380 kPa has been measured. From these results and additional measurements of the refractive index of Ne near 54.4 K, 83.8 K and 161.4 K, the differences ($T - T_{90}$) between the thermodynamic temperature T and its approximation T_{90} have been determined. From the refractive index measurements, the second density virial coefficient of helium and neon were estimated and in good agreement with the *ab initio* calculations.

RIGT measurements at 273.16 K in He and Ar have started using the apparatus previously used for the determination of the Boltzmann constant. A pressure vessel suitable for measurements up to pressures as high as 7 MPa has been constructed. RIGT thermometry over the temperature range 235 K and 430 K with a quasi-spherical copper resonator will start in late 2021 or early 2022.

List of publications

P. Czachorowski *et al*, Second virial coefficients for ^{4}He and ^{3}He from an accurate relativistic interaction potential (2020) Phys. Rev. A **102** 042810
https://doi.org/10.1103/PhysRevA.102.042810

C. Gaiser *et al*, Thermodynamic temperature data from 30 K to 200 K (2020) *Metrologia* **57** 055003 https://doi.org/10.1088%2F1681-7575%2Fab9683

B. Gao *et al,* Measurement of thermodynamic temperature between 5 K and 24.5 K with single-pressure refractive-index gas thermometry (2020) *Metrologia* **57** 065006
https://doi.org/10.1088/1681-7575/ab84ca

D. Imbraguglio *et al*, Comparison of ITS-90 realizations from 13 K to 273 K between LNE-CNAM and INRIM (2020) *Measurement* **166** 108225
https://doi.org/10.1016/j.measurement.2020.108225

G. Garberoglio and A. H. Harvey, Path-integral calculation of the fourth virial coefficient of helium isotopes, *J. Chem. Phys.* **154** 104107 (2021) https://arxiv.org/abs/2101.02624 (doi: 10.1063/5.0043446)

O. M. Hahtela *et al*, Coulomb Blockade Thermometry on a Wide Temperature Range *CPEM 2020 Proceedings* https://doi.org/10.1109/CPEM49742.2020.9191726 published

C. Pan *et al*, Direct comparison of ITS-90 and PLTS-2000 from 0.65 K to 1 K at LNE-CNAM, *Metrologia*, **58** (2021), 025005, https://doi.org/10.1088/1681-7575/abd845

A. Peruzzi *et al,* Survey of subrange inconsistency of long-stem standard platinum resistance thermometers (2021) *Metrologia* **58** 035009 https://doi.org/10.1088/1681-7575/abe8c1

C. Pan *et al,* Acoustic measurement of the triple point of neon T_{Ne} and thermodynamic calibration of a transfer standard for accurate cryogenic thermometry (2021) *Metrologia* **58** 045006 https://doi.org/10.1088/1681-7575/ac0711

D. Madonna Ripa *et al*, Refractive index gas thermometry between 13.8 K and 161.4 K (2021) *Metrologia* **58** 025008 https://doi.org/10.1088/1681-7575/abe249