Publishable Summary for 18SIB07 GIQS
Graphene impedance quantum standard

Overview

The aim of this project was to enable an economically efficient traceability of impedance quantities to the defining constants of the revised International System of Units (SI). This will simplify the calibration support which European and other National Metrology Institutes (NMIs) provide to the electronics industries. To meet these goals the partners have successfully developed high-quality graphene-based QHE impedance standards and improved the performance and the working range of digital- and Josephson-impedance bridges. These pieces of equipment are easier to operate and the provided Good Practice Guide will allow the users a simpler traceability of the impedance units to the quantum Hall effect with high metrological accuracy. The developed cryogenic system components for operating the quantum devices will help to simplify the challenging requirements of impedance measurements in the future.

Need

Electronic components rely, for their international competitiveness, on the application of mutually agreed measurements during their production and use. Electrical impedance (capacitance, inductance) is a quantity in this field that is practically of equal importance to voltage and resistance. It is more important than current, as the sensors, which are used in numerous contexts either rely on resistance or, more often, on contactless capacitive methods. For voltage and resistance, the “gold standards” of traceability, namely the Josephson effect (JE) and the quantum Hall effect (QHE), have been used in major NMIs for a long time. Impedance standards on the other hand still require many calibration steps with complicated measurement setups to trace them to the QHE.

At present, only a very few quantum standards are in use outside of the NMIs due to the high investment, high operational costs, and complexity of use. The quantum traceability of the impedance unit, the farad, would clearly benefit if an economically viable route was established. However, the complex calibration chains from the QHE to different capacitance and inductance values only exist in some of the largest NMIs. This was not acceptable and a shorter and simpler traceability chain of impedance to quantum standards, which is available and affordable for all NMIs, calibration centres and industries (e.g., automotive and mobile electronics), was clearly needed.

At this point, the need to utilise graphene came into play: its potential for metrology was understood almost immediately, because in graphene the QHE can exist at much lower magnetic fields (below 6 T) and at higher temperatures (above 4 K) than in conventional systems. The fundamental constant realisation for the DC QHE has been simplified by using graphene. The realisation of the corresponding fundamental constant for the AC units of impedance, i.e. capacitance and inductance, has also benefited from graphene with its much less demanding operational margins with respect to temperature and magnetic field. In addition, simpler and more flexible AC instrumentation had to be further developed, optimised and adapted for the use of graphene devices that are directly operated in the AC regime, thus avoiding a troublesome DC resistance to AC impedance transfer procedure.

Objectives

The overall objective was to combine novel digital impedance measurement bridges with the QHE material graphene in a simplified cryogenic environment. This has provided European NMIs, calibration centres and industry with the technology that is needed to enable the practical realisation of electrical impedance units (ohm, farad, henry) in the revised SI.

The specific objectives of the project were:

1. To optimise and to tailor graphene material and graphene devices in order to improve the understanding of the graphene AC quantised Hall effect (AC-QHE), as the basis for the traceability of impedance units to the QHE at temperatures of 4 K or higher in magnetic fields that are as low as possible - at most 6 T.
2. To advance digital bridges for the capacitance range from 10 pF to 10 nF at frequencies up to 100 kHz, and to develop an impedance bridge working with spectrally pure Josephson voltages up to 50 kHz in the entire complex plane.
3. To combine graphene devices with a Josephson impedance bridge (with a target uncertainty below 0.01 µΩ/Ω), and with a full digital bridge for simplified operation, (with a target uncertainty in the 0.1 µΩ/Ω range), in order to provide traceability for capacitance to the QHE.
4. To develop and investigate a cryo-cooler system hosting the superconducting Josephson device and the graphene device, both operating at AC and serving as the core element of a quantum resistance and impedance standard in the revised SI.
5. To facilitate the take up of the technology and measurement infrastructure developed in the project by the measurement supply chain (e.g. graphene manufacturers), standards developing organisations and end users (e.g. NMIs and calibration centres as well as the European Commission's Graphene Flagship).

Progress beyond the state of the art

Fabrication of graphene-based impedance quantum standards: The potential of graphene devices for use as impedance standards has been validated as they needed optimisation with regard to the special AC conditions which went beyond the goals of EMRP JRP SIB51 GraphOhm. Within this project, the consortium developed graphene devices which are stable in the long-term and which are optimised for AC applications for use in impedance metrology with an accuracy of 10^{-8}.

Advanced digital bridges: Classical transformer-based bridges for impedance calibration are limited in their frequency range, difficulty and they are lengthy to handle. The partners developed programmable, reconfigurable bridges that are usable over a large frequency range, and which go beyond the goals of the EMRP JRP SIB53 AIMQuTE. Within the project digital bridges were developed for the capacitance range from 10 pF to 10 nF at frequencies up to 100 kHz, and an impedance bridge was developed working with spectrally pure Josephson voltages up to 50 kHz in the entire complex plane.

Traceability of the capacitance unit: The combination of Josephson full digital impedance bridges for the realisation of a graphene-based quantum impedance standard was not attempted before. In this project for the first time a prototype of an impedance quantum standard based on graphene was planned with a digital impedance bridge and a state-of-the-art dual Josephson impedance bridge with a target accuracy of 10^{-8}.

Cryo-cooler system for graphene-based impedance measurements: Josephson voltage standard devices are typically very sensitive to external magnetic fields, whereas magnetic fields are indispensable for QHE devices. Therefore, the combination of both elements in one cryostat was not yet available. In this project, an experimental feasibility study on the operation of a graphene quantum Hall device and Josephson voltage standards in the same cryogen-free cryostat was carried out.

Results

To optimise and to tailor graphene material and graphene devices in order to improve the understanding of the graphene AC quantised Hall effect (AC-QHE), as the basis for the traceability of impedance units to the QHE at temperatures of 4 K or higher in magnetic fields that are as low as possible - at most 6 T.

A key for the fabrication of reliable resistance standards is the quality of the basic graphene layer. This was accomplished by epitaxial growth on silicon carbide substrates in three partner institutes. The resulting graphene layers consisted of homogenous monolayer graphene over areas, large enough for the fabrication of quantum Hall devices up to the mm range. The fabrication process of quantum Hall devices was optimised and has led to mechanically stable ohmic contacts with low resistance values of a few milliohm. Another key feature was the successful application of a molecular post-growth doping technique by which the electron density was tuned to values of 10^{11} cm^{-2}. This has allowed working points for the QH resistance measurements at low magnetic fields of $B < 6$ T. The quality of the fabricated quantum Hall devices was tested by dc-quantum

Hall measurements in partner labs in Europe and Asia which also proved the robustness of the devices and the long-term stability of their properties. Investigations at the BIPM (Bureau International des Poids et Mesures), a stakeholder of the GIQS project, testified that a graphene-based QHR standard agrees with the nominal quantised resistance value of a conventional GaAs standard with an accuracy of $(1\pm3)\times10^{-9}$ at a magnetic field of 5 T and a temperature of 4.2 K. This has represented an excellent confirmation of the high quality of the devices produced and it supports the preparations of BIPM for the transition from GaAs to graphene-based QHR reference standards.

In the next step and for the first time in quantum metrology further optimisation steps were tackled to improve the AC properties of the graphene devices. These included the fabrication of a special sample holder dedicated to ac experiments as well as adapting the device and the Hall bar geometry to ac currents. The validation of the fabricated ac-QH devices in the partner labs resulted in a relative accuracy of a few parts in 10^{-8} for the reproduction of the nominal quantised resistance value which is comparable to conventional galliumarsenide-based impedance standards. The successful investigations by the partners also showed that these ac-QH standards were well suited for the evaluation of the impedance bridges. The project results impressively confirm that graphene-based QH devices can serve as the basis for the traceability of impedance units to the QHE at temperatures of 4.2 K and at magnetic fields below 6 T and that the objectives were fully achieved.

To advance digital bridges for the capacitance range from 10 pF to 10 nF at frequencies up to 100 kHz, and to develop an impedance bridge working with spectrally pure Josephson voltages up to 50 kHz in the entire complex plane.

Different types of digital impedance bridges were upgraded and evaluated in detail in the labs of the project partners. The evaluation and optimisation of the bridges has led to lower uncertainties, higher stability and faster balancing of the bridges. The capacitance calibration was carried using a 10 nF capacitance standard which was directly linked to a 12906 Ω resistance standard with a low total uncertainty of 7×10^{-8}. The down scaling of the capacitance scale to a 10 pF capacitor was attained by ratio measurements. For the high-frequency upscaling from 1.233 kHz to 100 kHz a high-frequency digitally-assisted bridge was used, and the evaluation was carried out by means of the calibrated 10 pF capacitor. It turned out that the target uncertainty of 10^{-5} could be surpassed with a total uncertainty budget for the highest frequencies of better than 7.3×10^{-6} (value at 100 kHz).

Further progress could be achieved by improving and evaluating the dual Josephson impedance bridges (DJIB) which were set up in two labs (METAS and PTB). This type of bridge is taking advantage of the spectral purity, the low noise level and quantum-based accuracy of the pulse-driven Josephson voltage standards used for the potential definition within the impedance bridge. The frequency range of this versatile instrument spans from about 50 Hz to 60 kHz. The comparison to conventional impedance bridges showed only a very small deviation of $(2\pm7)\times10^{-9}$ for the link from a 10 nF capacitance standard to the quantum Hall resistor. This has exceeded the originally targets and has shown that DJIBs can approach uncertainties of traditional impedance bridges. Moreover, the full automation makes it a user-friendly tool for almost all kinds of impedance calibrations. The objective was fully achieved.

To combine graphene devices with a Josephson impedance bridge (with a target uncertainty below 0.01 µΩ/Ω), and with a full digital bridge for simplified operation, (with a target uncertainty in the 0.1 µΩ/Ω range), in order to provide traceability for capacitance to the QHE.

During the project digitally assisted, fully digital and Josephson impedance bridges were extended to integrate a quantum Hall resistor in a so-called triple series connection. Using this it has been possible to link any kind of impedance to the quantised Hall resistance. The unique features of the quantised Hall resistance in a multiple series connection allowed us to dispense with a combining network and led to a compact and simple design of the whole setup. Moreover, the low noise of a quantised Hall resistor has reduced the measurement time compared to resistance standards at room temperature. The combination of graphene QHR devices in all kinds of impedance bridges used within the project worked out successfully without unexpected problems at five institutes of the consortium. No additional noise sources were observed in this complex setup. This was confirmed by the investigation of the statistics by means of Allan deviation measurements which were within the theoretical expectation. With all these efforts it is now possible to link any kind of impedance to the QHR over a broad range of frequencies and with automated systems. For example, the link of capacitance standards in the range of 8 nF to 10 nF could be linked to the QHR with uncertainties (k=1) below 1 part in 10^7 for digital bridges and a few parts in 10^8 for Josephson impedance bridges.

One important type of measurements with these combinations is the evaluation of QHR devices themselves to provide feedback to the device manufacturing groups. These evaluations were done successfully within the targeted uncertainties at all involved institutes. A mutual validation of a digital and a Josephson impedance bridge was performed within a Researcher Mobility Grant (RMG) by evaluating the discrepancies between the calibrations of the standards obtained with the two bridges. This successful comparison agreed within the statistical uncertainties - a few parts in 10^8. The objective was fully achieved.

To develop and investigate a cryo-cooler system hosting the superconducting Josephson device and the graphene device, both operating at AC and serving as the core element of a quantum resistance and impedance standard in the revised SI.

The developed cryo-cooler systems have provided a user-friendly tool for the realisation of the impedance units. This is based on one hand by the implementation of a Josephson-voltage-source impedance bridge for a simplified operation and on the other hand by using developed graphene-based resistance standards which allow working points at low magnetic fields (< 5 T) and high temperature (4.2 K) and thus, the use of a closed cycle cryocooler. For the first time worldwide, this project has dedicated itself to this task which lies in the combination of both features in one cryo system. The resulting challenges were addressed by two independent approaches in two institutes. The remaining very low noise level (Allan variance below 1 nΩ/Ω after 30 min) demonstrated that it is, indeed, possible to use the developed system for metrology-grade quantum Hall resistance measurements with an accuracy better than 10^{-9} which is the standard metrological precision of the usually used liquid helium cryostats in NMI's. More optimisation steps were undertaken, e.g. improving thermal coupling of the Josephson-voltage-source and wiring which were highly beneficial for the performance of the JVS under realistic experimental conditions. By proper positioning and shielding of the Josephson-voltage-source an effective protection again stray fields of the magnet was achieved. Even at high magnetic fields (10 T), which are stronger than necessary to achieve quantisation of the graphene resistance standard, an undisturbed function of the Josephson voltage source is guaranteed. This project has successfully developed a cryo-cooler system and the investigations show that all specific problems have been completely solved. This will allow a successful operation of a Josephson voltage standard combined with a graphene-based quantum Hall standard in a single cryogenic system serving as the core element for the realisation of the resistance and impedance unit in the revised SI. Only the final simultaneous test of the cryostat including all components will need to be performed at a later time, otherwise the objective was achieved.

Impact

To promote the uptake of the progress in the field of impedance metrology and epitaxial graphene fabrication technology generated in this project, as well as to share insights generated throughout the project, the results were shared broadly with scientific and metrology end-users.

Twelve papers in international peer-reviewed journals, a book chapter and two PhD theses were published and submitted, respectively. 33 presentations were held at conferences, including those associated to the Graphene Flagship, the Conference on Precision Electromagnetic Measurements, the International Metrology Conference, the IMEKO TC-4 conferences, and the URSI General Conference. Information on the GIQS activities were spread to regulatory bodies. In the past years the EURAMET committee TC-EM (Electricity and Magnetism) was regularly informed about the progress of the GIQS project at the annual contact person meeting as well as at the 23rd TCEM meeting of the Asian Pacific Metrology Programme (APMP). Technical committees of GULFMET, COOMET and SIM were also kept informed. The CCEM has adopted one of the Newsletter issues as a working document.

19 lab training activities were carried out for scientists from the consortium and for external audiences, among them 8 presentations at university seminars. The consortium disseminated project results in many other ways, e.g. PhD theses and articles in trade journals.

Five newsletter issues which summarise the goals and results of the project were produced and distributed to stakeholders. To inform stakeholders, the scientific community and interested end users about news from the project in the short term, four online platforms were created. Next to the project's homepage, GIQS groups exist on LinkedIn and ResearchGate. 15 tutorial videos on the latest research results in the field of graphene fabrication for quantum metrology and impedance measurements using the developed bridges are available from the GIQS YouTube channel.

To learn more about the needs and expectations of the stakeholders the consortium has proactively launched a survey among potential stakeholders. In a subsequent joint stakeholder meeting the consortium presented the results in the work packages and a discussion about further activities was started. In Winter 2020 a dedicated workshop for stakeholders and collaborators was organised online.

Impact on industrial and other user communities

This project established a new primary standard of impedance. At present, industrial and other user communities rely on a long chain of calibrations, originating at the BIPM or few large NMIs, and extending over smaller NMIs, and/or commercial calibration service providers to eventually reach the end user. This lengthy chain has been shortened with a simpler, cheaper and easier to operate primary standard. The impact on industrial and other end users will gain its full thrust after an initial post-project phase when the knowledge and know-how created in this project has been taken up by instrumentation companies and converted into marketable solutions e.g a digital impedance bridge based on conventional electronics. The early impact will be on industrial instrumentation manufacturers and large-scale calibration service providers, which will be able to use the enhanced calibration quality enabled by the new primary impedance quantum standard to provide scientifically sound services with improved performance, reliability and cost efficiency.

Impact on the metrology and scientific communities

At present many small NMIs obtain traceability for their national resistance and capacitance standards from other NMIs or from BIPM due to the high acquisition and operational costs of primary quantum Hall systems. For example, from 2013 – 2017, BIPM issued 159 certificates for resistance and 127 for capacitance for 36 NMIs. Eighteen of them were from Europe. Therefore, at the NMI level, one of the major impacts of the project will be to provide a direct and user-friendly realisation of impedance units in the "revised SI" at the "point of calibration", thereby relaxing the NMIs' dependence on BIPM, and releasing BIPM's resources for more important research tasks.

To create early impact in the scientific communities, research papers have been published in high impact peer-reviewed journals. Dissemination workshops will also be organised for different target groups.

As explained in the draft mise en pratique for the ampere and other electrical units in the revised SI, the unit farad can be realised by comparing the quantised Hall resistance to the impedance of the unknown capacitance using, for example, a quadrature bridge. Development of digital solutions, e.g. digital conventional bridges, for such a primary unit realisation will be one of the main impacts of this project. The graphene-based quantum Hall resistance (QHR) device is also valuable in the realisation of other electrical units of the revised SI, such as the unit ampere that can be realised *"by using Ohm's law, the unit relation A = V/Ω, and using practical realisations of the SI derived units volt V and ohm Ω, based on the Josephson and quantum Hall effects, respectively"*. For those NMIs who decide to implement the new impedance quantum standard, such direct access to the primary capacitance realisation will enable them to claim improved CMCs in this field.

The BIPM has publicly recognised the relevance of the approach proposed by the GIQS project by performing dc electrical metrology experiments with graphene devices developed within the project.

Impact on relevant standards

This project will create impact as an impedance calibration system based on a quantum standard is now available. How this new primary quantum standard, and especially its possible use outside of authorised NMIs, will impact the international measurement system and written standards need to be discussed by the relevant bodies of the international measurement system. However, this project has actively contributed to key

European and international committees (e.g. Consultative Committee for Electricity and Magnetism (CCEM)) to spread the required information and knowledge.

In this context, the most important regulatory document is EN ISO/IEC 17025, “General requirements for the competence of testing and calibration laboratories”, which defines in general terms the technical requirements for good practice calibrations in section 6.5 (2017). Furthermore, calibration is an important and recurring subject in the EC directive MID 2004/22/IEC on measuring instruments. National laws and directives in the EU member states concerning the units of measurements and how they are to be implemented are relevant law whose simpler, better, and more cost-effective compliance will be targeted by the developments in this project.

Longer-term economic, social and environmental impacts

Achieving the objectives of this project will respond to the need for a shorter traceability chain to quantum impedance standards by using the huge metrological potential of graphene. It will also preserve and strengthen Europe’s lead in the metrological applications of the QHE in graphene and in digital impedance metrology.

The European calibration services market was estimated to be worth $1.55 billion in 2018, with more than 40 % of the total being for electrical calibrations, which include instrument calibrations for radiation dosimetry, medical diagnostics and treatment, smoke detectors, devices for environmental monitoring, semiconductor wafer characterisation, etc. Even a relatively small improvement in the accuracy for the end users, as a result of the shortened calibration chain targeted in this project, will generate a very significant amount of economic benefit for the EU. In the technology sector, where the benefit-to-cost ratio of electrical measurements is amongst the highest, the development of new types of devices, sensors, and measurement methods with faster speed, higher sensitivity, and lower energy consumption, will become possible.

The long-term environmental impact of this project will be indirect, yet strong; many measurement techniques, be they environmental, medical or scientific, make use of transducing elements based on capacitance. Improving the traceability to capacitance will improve the sensitivity and reproducibility of such measurement techniques, leading in turn to improved data quality which will lead to several benefits: more efficient engines, a reduction in exhaust gases, the measurement of polluting particulates, and to an improvement in electrical impedance spectroscopy for geophysics, to name only some. The political benefit of the research will be that a high-end primary standard will become attainable for almost every metrology or calibration laboratory. Proliferation of primary quantum standards is one of the key aims of international metrology and it allows all countries to interact on an equal basis.

List of publications

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[3] M. Marzano, M. Ortolano, V. D’Elia, A. Müller, and L. Callegaro, A fully digital bridge towards the realization of the farad from the quantum Hall effect, Metrologia 58, 015002 (2021). https://doi.org/10.1088/1681-7575/abba86

[4] S. Bauer, R. Behr, R. E. Elmquist, M. Götz, J. Herick, O. Kieler, M. Kruskopf, J. Lee, L. Palafox, Y. Pimsut, and J. Schurr, A Four-Terminal-Pair Josephson Impedance Bridge Combined with a Graphene Quantized Hall Resistance, Meas. Sci. Technol. 32,065007 (2021), https://doi.org/10.1088/1361-6501/abcff3

[5] M. Kruskopf, S. Bauer, Y. Pimsut, A. Chatterjee, D. K. Patel, A. F. Rigosi, R. E. Elmquist, K. Pierz, E. Pesel, M. Götz, and J. Schurr, Graphene quantum Hall effect devices for ac and dc electrical metrology, IEEE Transactions on Electron Devices 68, 3672 (2021). https://doi.org/10.1109/TED.2021.3082809

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[7] D.-H. Chae, M. Kruskopf, J. Kucera, J. Park, N. T. Mai Tran, D. B. Kim, K. Pierz, M. Götz, Y. Yin, P. Svoboda, P. Chrobok, F. Couëdo, and F. Schopfer, Investigation of the stability of graphene devices for quantum resistance metrology at direct and alternating current, Meas. Sci. Technol. 33, 065012 (2022). https://doi.org/10.1088/1361-6501/ac4a1a

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[9] Momeni Pakdehi, Davood., Optimization of Epitaxial Graphene Growth for Quantum Metrology, PhD thesis, 2020, Gottfried Wilhelm Leibniz Universität Hannover, Germany, https://doi.org/10.15488/10201

This list is also available here: https://www.euramet.org/repository/research-publications-repository-link/

<table>
<tr><td colspan="2">Project start date and duration:</td><td>1 June 2019, 36 months</td></tr>
<tr><td colspan="3">Coordinator: Klaus Pierz, PTB   Tel: +49 531 592 2412   E-mail: klaus.pierz@ptb.de
Project website address: https://www.ptb.de/empir2019/giqs/home/</td></tr>
<tr><td>Internal Funded Partners:
1. PTB, Germany
2. CMI, Czech Republic
3. INRIM, Italy
4. LNE, France
5. METAS, Switzerland
6. RISE, Sweden
7. VTT, Finland</td><td>External Funded Partners:
8. CNRS, France
9. NIMT, Thailand
10. POLITO, Italy</td><td>Unfunded Partners:
11. KRISS, Korea, Republic of</td></tr>
<tr><td colspan="3">RMG1: INRIM, Italy (Employing organisation); PTB, Germany (Guestworking organisation)</td></tr>
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