D4.2| Stand-alone unit

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Abstract

The SUPERTED project has been focused on the development of electromagnetic radiation sensors based on the Giant Thermoelectric Effect which appears when superconducting (S) and ferromagnetic insulating (FI) materials are combined. Many difficult technical challenges have been faced and solved, from the realization of stable and reproducible FI/S junctions, to the design and fabrication of sensors and appropriated read-outs.

We believe it is very important to make all this knowledge accessible for researchers, whatever they background, so that they can push the development of this technology and its applications further with their own contributions. As part of this knowledge transfer effort, we explain in this document how a basic sensing set up is made from scratch.

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We have divided the document in four main sections. In the first chapter we describe the working principle of the sensors, i.e. how the Giant Thermoelectric Effect can be used as a basis for this type of devices. In chapter 2 we describe the different processing steps that allow to build the sensors. In chapter 3 we describe the devices themselves (and, in particular, versions adapted for detection of radiation in the X-ray and THz ranges, respectively), with a focus on the parameters that can be optimized. Finally in chapter 4 we describe the integration into the working (cryostatic) environment, and the read-out required to extract the signals.

Since the potential readers may have very different backgrounds, we have tried to make this an easy reading, as intuitive as possible text, although inside it we point out towards the most relevant scientific publications produced within the project, which contain much more detailed discussions about the specific topics.

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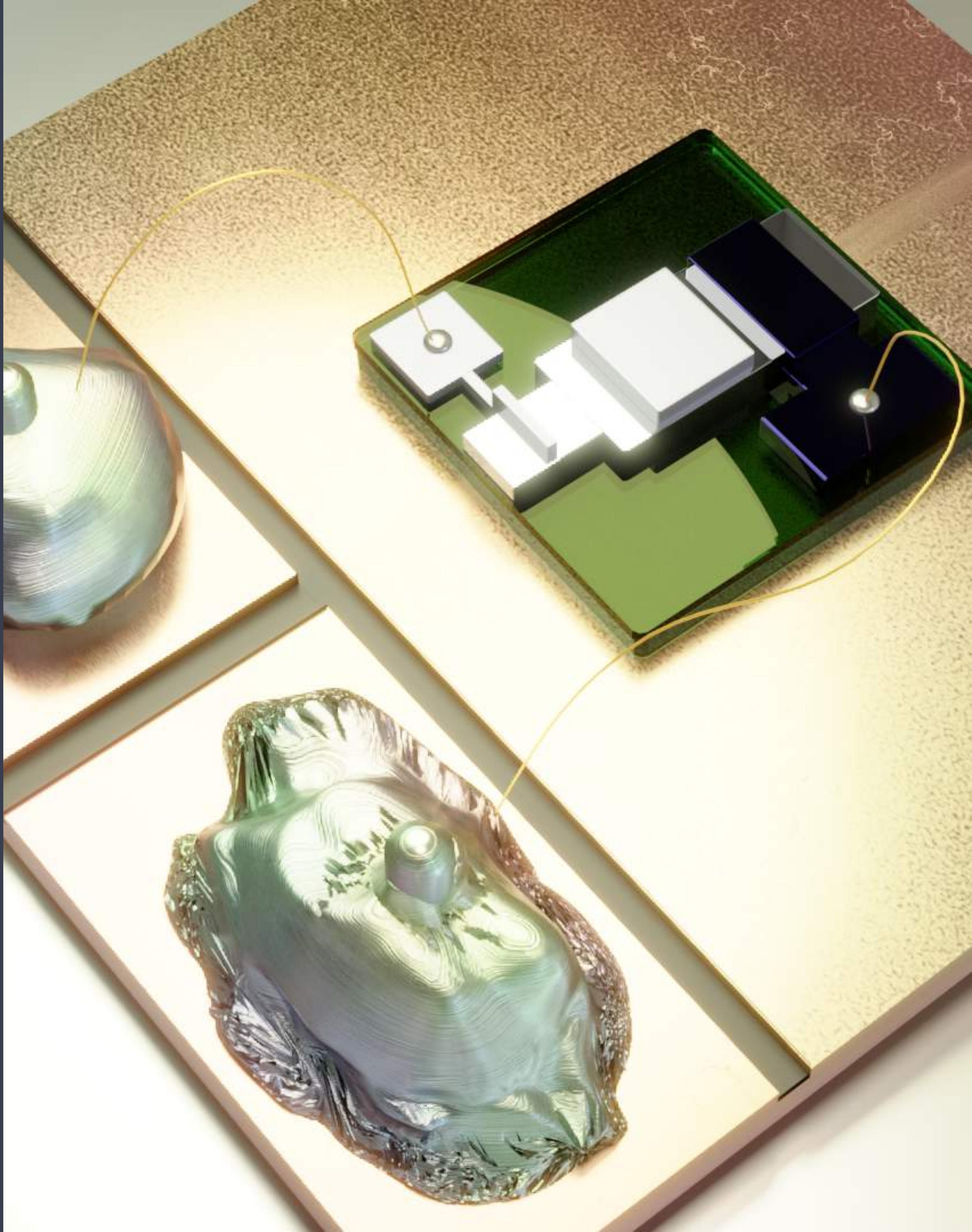
SUPERTED

STAND-ALONE UNIT

A Visual Introduction to radiation sensors based on Giant Thermoelectric Effect



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FOREWORD

The discovery of a new physical phenomenon is an exciting moment in scientific research. It implies that we have gained a deeper insight about how our Universe works. This has been the case almost ten years ago, when some of the partners of the SUPERTED Consortium have discovered the Giant Thermoelectric Effect, which appears when certain materials are arranged in a specific manner.

Discoveries are also an opportunity to find ways of harnessing nature to our own advantage, through the development of new technologies. This is a complex process, which often requires contributions from people with many different backgrounds and skills. The timescale for developing a new technology can extend many years, and largely depends on the capacity of the stakeholders to gather and focus resources, but also on their ability to disseminate the knowledge and motivate other actors along the value chain to get involved and boost the new technology further with their contributions.

The Framework Programmes for Research and Technological Development set in place by the European Commission have proven to be an extremely efficient catalyser for scientific discovery and technology development. By providing sufficient level of funding to selected projects, they allow constituting multidisciplinary consortiums that work in a coordinated manner, drastically easing the exchange of information and training, and speeding the overall process.

In the case of the SUPERTED project, this focused effort has allowed developing high performance sensors whose functioning is underpinned by the Giant Thermoelectric Effect. While it was obvious by the beginning of the project that some scientific applications, like X-ray Spectroscopy and Astrophysics in the THz/GHz range, could directly benefit from the use of these devices, we also thought that it was important to devote part of the effort to explore other possible uses of this technology and, most importantly, to prepare the outcome of the project so that next generations of researchers could use it and build on top of it.

Thinking of this knowledge transfer process, we have worked under the hypothesis that it is likely that new actors may not be as lucky as we have been. The EC funding has allowed us to access a very large and powerful set of resources, and to put together a world-class team covering many disciplines. Thanks to this, we have been able to develop new materials, design and build new structures for the sensors, implement read-out schemes in complex (cryogenic) environments and test the devices in realistic application set ups. But for a researcher from a particular area, with access to a more limited set of resources and with knowledge of a particular topic, it may be a real challenge to reproduce at least part of the outcome of the SUPERTED project, that normally would be the starting point of their research. Actually, even the first step of understanding all the work that has to be done, and linking with other scientist who can collaborate, will already be a major effort.

In order to compensate for this situation, we scheduled specific activities in the project which aimed at wrapping up, structuring and presenting in an easy to digest manner the outcome of the project. In a nutshell, the idea behind these activities was to build the simplest possible working versions of the sensors, which were named "stand-alone" prototypes, and describe the process of building them.

This document summarises this effort. It has been conceived as an information hub, where readers from any background and technical level will find (hopefully easy reading) introductions to the different concepts and topics together with links (in the blue shadowed areas) to the most relevant elements of the scientific bibliography and technical reports which have been produced in SUPERTED, which is where the outcome of the project has been presented exhaustively. Expert researches are encouraged to explore further these references in order to gain an in-depth understanding of the science and technology behind these new sensors.

Another important element of information in this document is provided by the links to the members of the SUPERTED consortium who have been in charge of the different activities. They are willing to get in touch and collaborate with anybody who could be interested in continuing to explore this exciting new technology.

In these blue shadowed areas you will find references toward relevant technical documents, including project deliverables and scientific publications.

To start with, an also general, but technically detailed, overview of the technology can be found in the review *Superconductor-ferromagnet hybrids for non-reciprocal electronics and detectors*, DOI: https://doi.org/10.48550/arXiv.2302.12732

ABOUT THE PROJECT

The SUPERTED project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No. 800923.

The project ran its activity from 1st September 2018 until 28th February 2023.

The members of the international Consortium where:

- **Jyväskylän yliopisto (University of Jyväskylä)**, from Finland.
- **Agencia Estatal Consejo Superior de Investigaciones Científicas (CSIC)**, from Spain.
- **Consiglio Nazionale delle Ricerche (CNR)** from Italy.
- **Centre National de la Recherche Scientifique (CNRS)** from France.
- **Bihurcrystal SL**, from Spain.
- **Advacam Oy**, from Finland.

JYVÄSKYLÄN YLIOPISTO
UNIVERSITY OF JYVÄSKYLÄ

CSIC
CONSEJO SUPERIOR DE INVESTIGACIONES CIENTÍFICAS

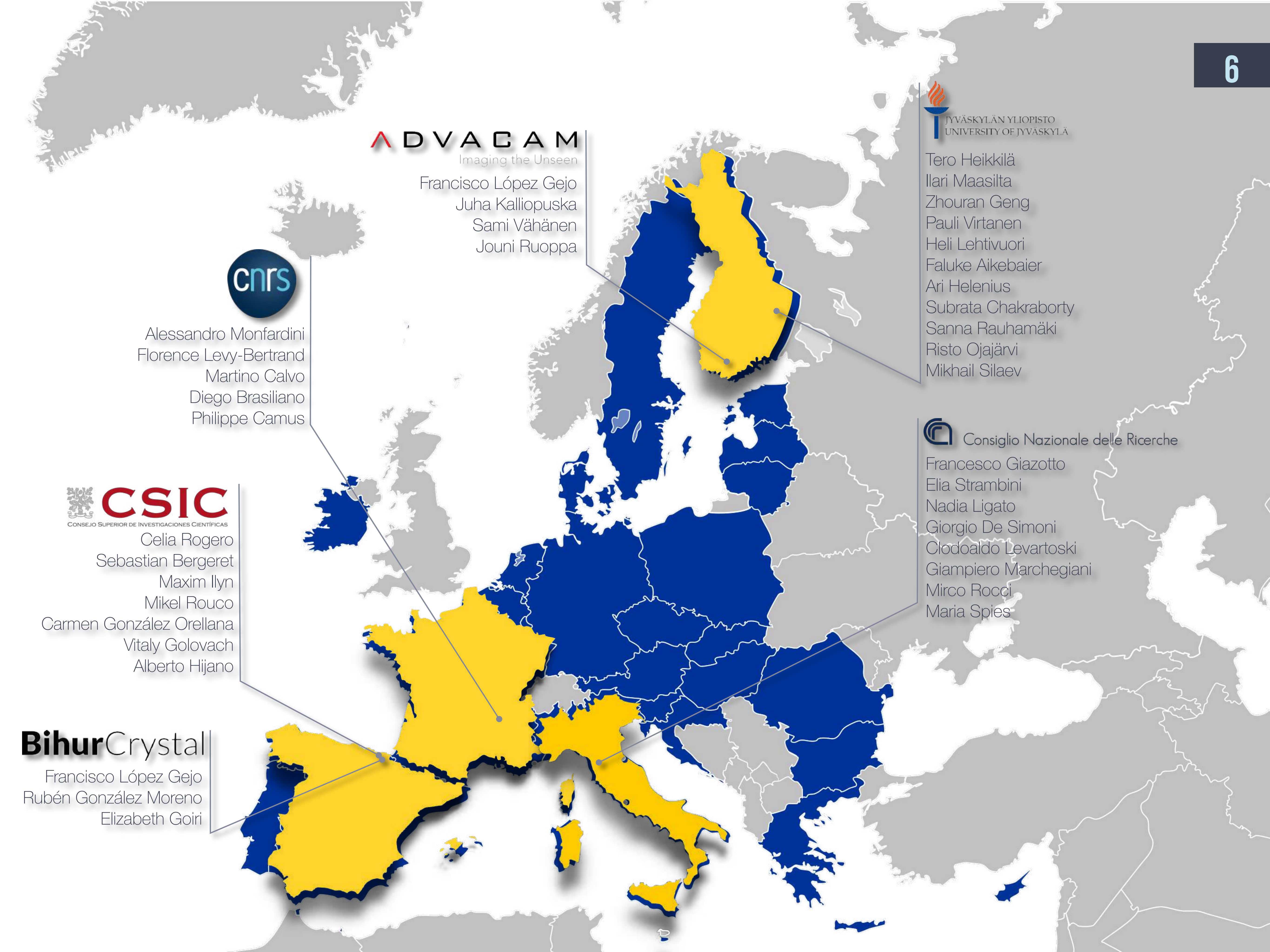
Consiglio Nazionale delle Ricerche

cnrs

BihurCrystal

ADVACAM
Imaging the Unseen

SUPERTED borrows its name from the animated television series about a teddy bear with superpowers. The character was created by Mike Young, as a way of helping his son to overcome the fear of the dark. We thought that maybe the high performance radiation sensors could help with that as well…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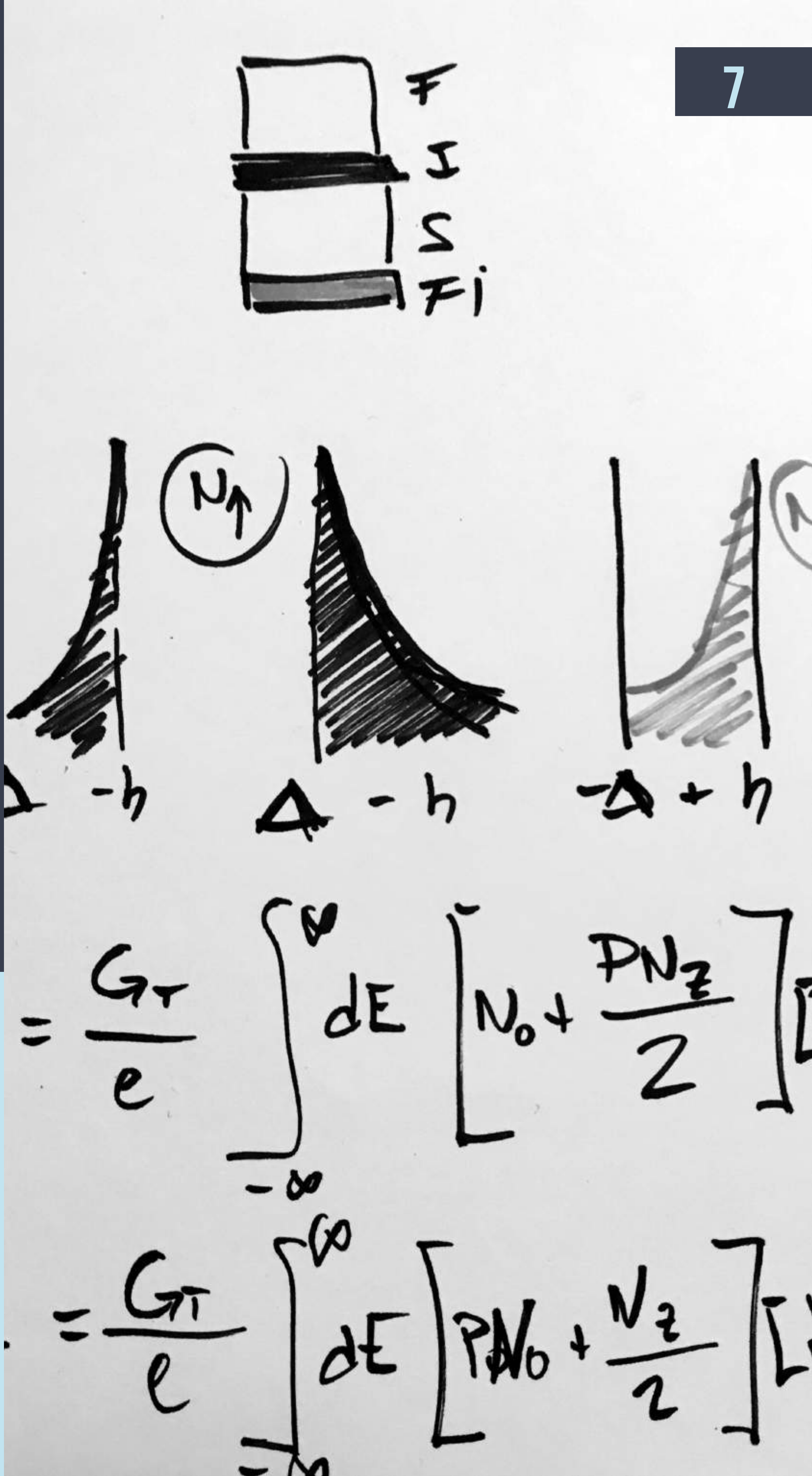
Rubén González Moreno
Elizabeth Goiri

CHAPTER 1

WORKING PRINCIPLE

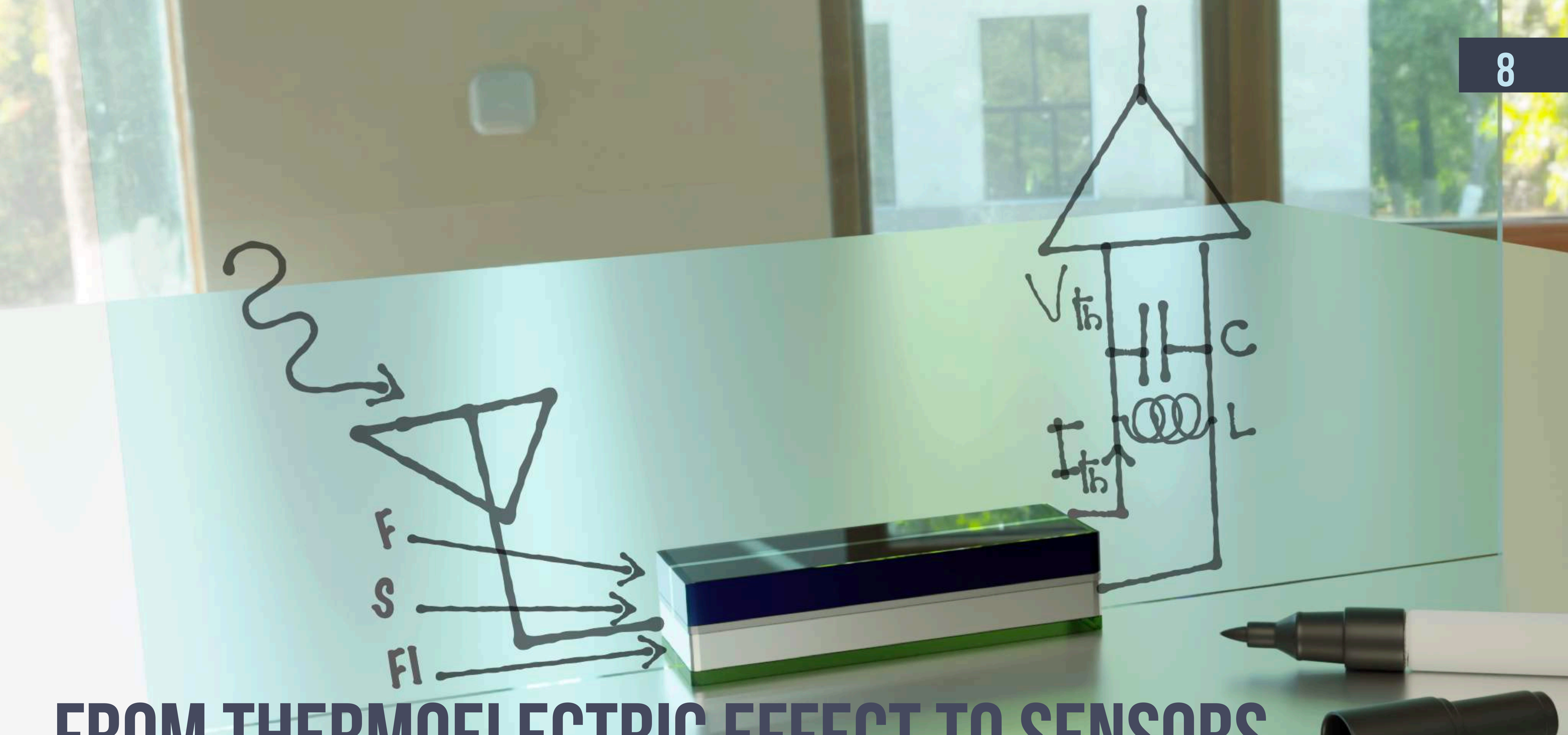
The harnessing of the Giant Thermoelectric Effect, and the design and optimisation of the sensors could not have been carried out without a deep understanding of the fundamental physics that underpins this physical phenomena.

The detailed description of the theoretical framework is a rather extensive exercise, which has indeed already been carried out in the scientific publications produced in the SUPERTED project. The aim of this chapter is to provide an initial approach to the subject, as intuitive as possible, and to point to the most relevant elements of this bibliography.



The development of the theoretical framework that allowed the description and study of the sensor properties in the SUPERTED project was mainly carried out in Work Package 2, and took place at University of Jyväskylä (Finland) and Centro de Física de Materiales (Spain). This work was led by:

- **Tero Heikkilä** (tero.t.heikkila@jyu.fi)
- **Sebastián Bergeret** (sebastian_bergeret@ehu.eus)



FROM THERMOELECTRIC EFFECT TO SENSORS

Sensors are devices that produce an output signal whose magnitude depends on the properties of a physical phenomenon and, therefore, can be use it to detect and characterise it. In practice, the vast majority of sensors are nowadays designed to produce electric signals, mainly because this allows a smooth integration with digital processing systems (i.e. computers). The core of a sensor must then be composed of a structure which allows for this conversion of the physical signal into an electrical signal. Such structure is usually know as a transducer.

Transducers must produce this conversion in a way where the output signal keeps its proportionality to the input signal, in a known way, over a range of signal amplitudes and characteristics as wide as possible (these are the dynamic range and bandwidth of the sensor). It is also desirable that the conversion allows discriminating small values of the input (this is the resolution of the sensor). Ideally, this discrimination must allow identifying the individual events related to the physical phenomenon which is being monitored (such an event can be, for instance, the impact of a single photon). The conversion process must take place as isolated as possible from other physical effects, as otherwise part of the output will correspond to contributions from these spurious phenomena (this is the noise of the detector).

While many different types of transducers exist, most of the available technologies meet the requirements listed above only up to a certain point. Very few can be considered as candidates for high-performance sensors, which are those with very large dynamic range and bandwidth, very high resolution and very low noise.

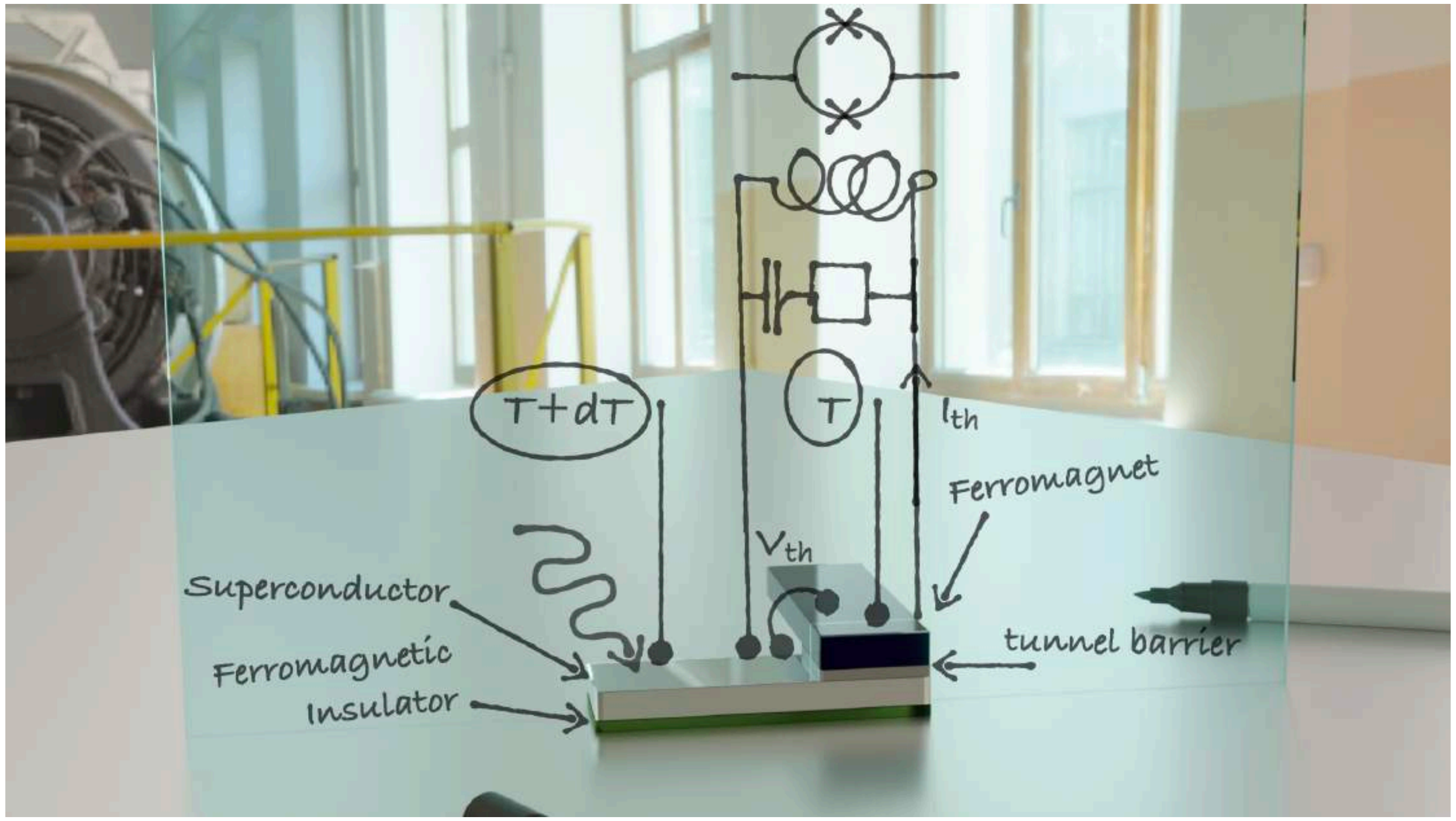
The sensors developed at SUPERTED project are based on the recently discovered Giant Thermoelectric Effect (*), which appears in a particular type of multilayered structures containing Ferromagnetic and Superconducting materials.

The fundamental description of these effect can be done using the formalism used for describing and studying the three main thermoelectric phenomena (Seebeck, Peltier and Thomson effects). It is well known that these mechanisms are enabled when the symmetry between positive and negative charge carriers (electrons and holes) is broken inside a material.

Naturally appearing thermoelectrical effects are generally weak, although the progress in the engineering of semiconductor materials has allowed obtaining artificially produced materials with higher responses. While superconducting materials share some (superficial) similarities with semiconductor materials (like, for instance, a quasiparticle transport strongly influenced by the presence of an energy gap), the magnitude of thermoelectric effects in superconductors is actually small, because in these materials the electron-hole symmetry is forced (by the charge neutrality).

(*) The seminal work, by several members of the SUPERTED consortium, where the Giant Thermoelectric Effect in FI/S junctions was first and deeply described can be found in *"Predicted Very Large Thermoelectric Effect in Ferromagnet-Superconductor Junctions in the Presence of a Spin-Splitting Magnetic Field"*, Phys. Rev. Lett. **112**, 057001 (2014). DOI: https://doi.org/10.1103/PhysRevLett.112.057001

An in depth review about this physical phenomena can be found in *"Thermal, electric and spin transport in superconductor/ferromagnetic-insulator structures", Progress Surf. Sci.* ***94****, 100540 (2019). DOI:* https://doi.org/10.1016/j.progsurf.2019.100540



FI/S structure used as a temperature transducer. If energy is supplied to the superconducting layer, it will increase its temperature by dT. The Giant Thermoelectric Effect will lead to the appearance of a voltage V_{th} between the superconducting and the ferromagnetic layers. If a circuit is established between these layers, this voltage will generate a current I_{th}.

This situation changes drastically when the superconductor is put in contact with a ferromagnetic material. This induces the appearance of a spin splitting field, which makes electrons with antiparallel spin to acquire a different amount of energy than electrons with parallel spin, and leads to the rupture of the hole-electron symmetry for each spin separately, while preserving the overall charge neutrality.

The breaking of the electron-hole symmetry causes a very large thermoelectric response: if two of the layers of the structure are placed at different temperatures, a relatively large electrical current will be established between them. This provides the fundamental basis for sensing devices which are able to carry out very fast and accurate measurements of the energy that induces the aforementioned changes in temperature (**). Such a transducer can be coupled to other elements which will be responsible for interacting with the environment and gathering the energy that will induce temperature changes. These elements can, for instance, be designed to interact with incoming photons and absorb at least part of their energy, effectively transforming the whole system into an electromagnetic radiation sensor.

Any transducer which measures changes in temperature is intrinsically affected by thermal noise, i.e., will also be sensitive to temperature changes induced by unwanted (and uncontrollable) energy inputs. From this point of view, the use of superconducting material turns convenient, because the superconducting state can only be reached in very low

(**) The theoretical study of an electromagnetic radiation detector based on the Giant Thermoelectric Effect can be found in the following publication: *"Thermoelectric Radiation Detector Based on Superconductor-.ferromagnet Systems"* Phys. Rev. App.. **10**, 034053 (2018). DOI: doi.org/10.1103/PhysRevApplied.10.034053 temperature environments, where the amount of available heat is scarce and the sources of noise are largely suppressed. This situation is actually advantageously exploited by other high-performance sensing technologies (*), also based on the use of superconducting materials, like the Transition Edge Sensors (TES) or the Kinetic Inductance Detectors (KID), but there are fundamental limitations why these other technologies cannot fully suppress the thermal noise.

The transducers of any of the aforementioned technologies are actually based on physical effects that modify the properties of an electrical currents or voltages which has to be supplied to the sensor in order to make it work. The circulation of these currents through the wiring constitutes a source of noise which, not only cannot be eliminated, but actually gets worse in complex devices such as sensor arrays, which are basic for many applications. This is not the case for the sensors based on the Giant Thermo-electric Effect, because in this case the current is actually generated by the transducer itself. This paves the way for the the construction of sensors, where contributions to noise due to heat dissipation are drastically reduced.

Radiation sensors based on temperature measurements will actually behave very differently depending of how they are configured:

- In Microcalorimeters, the system is tuned so that it produces an output signal and resets (gets ready for the next measurement) before the arrival of the next photon. The reset implies that the heat brought in by one photon is expelled out of the sensor (to a thermal bath). Such devices are suited for measuring the energy of incident particles.

- In Bolometers, the system recovery time is much longer than the interval between the arrival of two photons. In this case the device will be suitable for measuring a flux of radiation, typically from black-body emissions or radiation from the infrared range of the electromagnetic spectrum and beyond.

In principle, the configuration of a sensor (for working as micro-calorimeter or bolometer) will be done by design. The first element that will characterise the behaviour of the sensor is the choice of the absorber, which is very different depending on the type of incoming radiation. For the X-ray range, characterised by short wavelength and high energies, the absorber must be a material capable of stopping the photons in a short distance while for the THz range,

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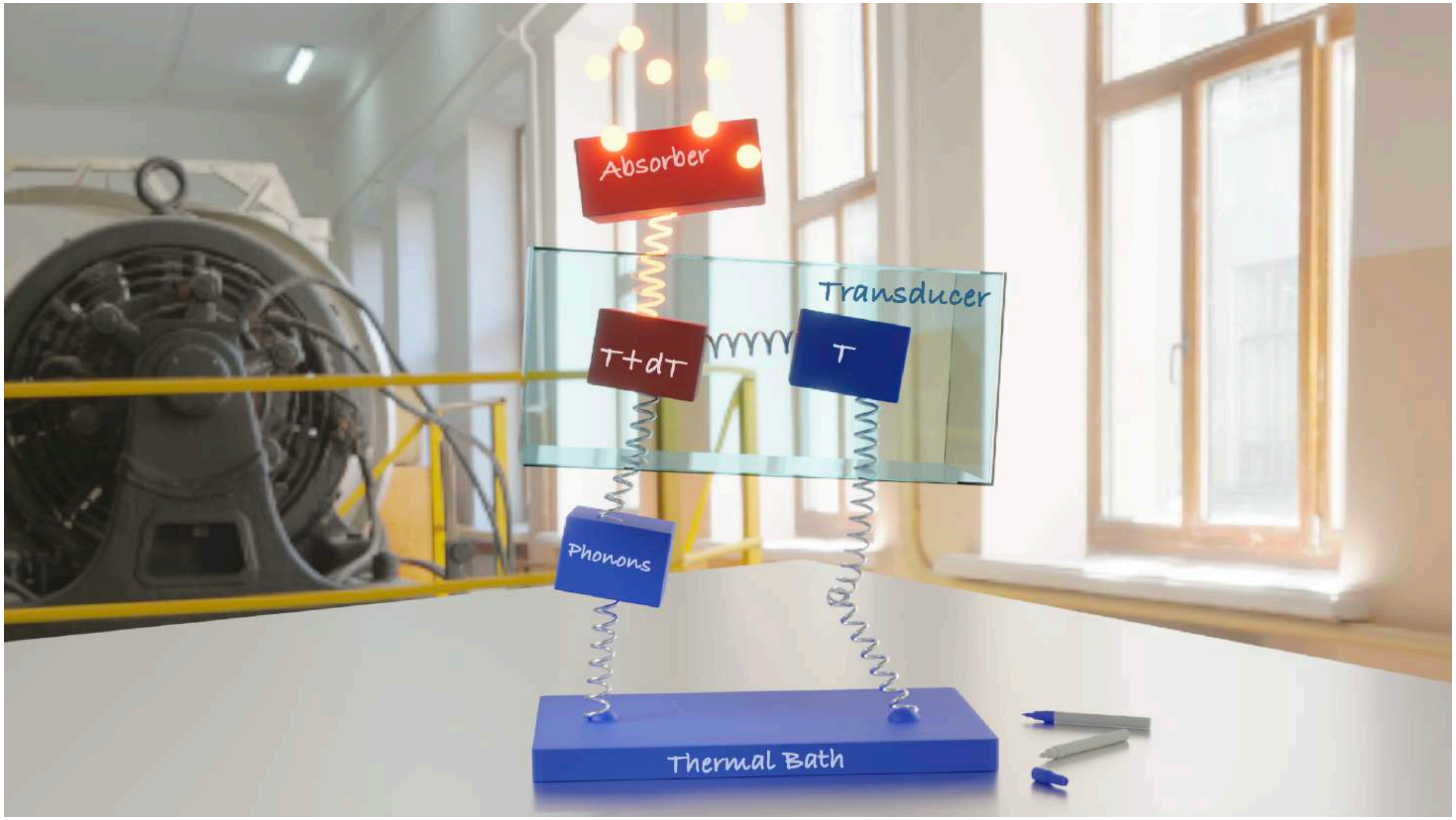
(*) An in depth study of other high-performance sensing technologies can be found in SUPERTED Deliverable 4.2. *"Technology Transfer Report".*

characterised by long wavelengths and low energies, the absorber must behave as an antenna that couples the energy of the electromagnetic radiation into the sensor. The absorbers must be engineered so that the heat flow follows the appropriate paths in the sensor, to minimize the part of the energy that is scattered without producing any output signal.

The same engineering of the heat flow must be carried out in the transducer. The parameters which play a role are the characteristics of the materials (specific heats) but also the geometrical features like the contact areas between the different elements. Care should be taken, however, as the same parameters play a role in some of the sources of noise. All these optimisations are explored in Chapter 3.



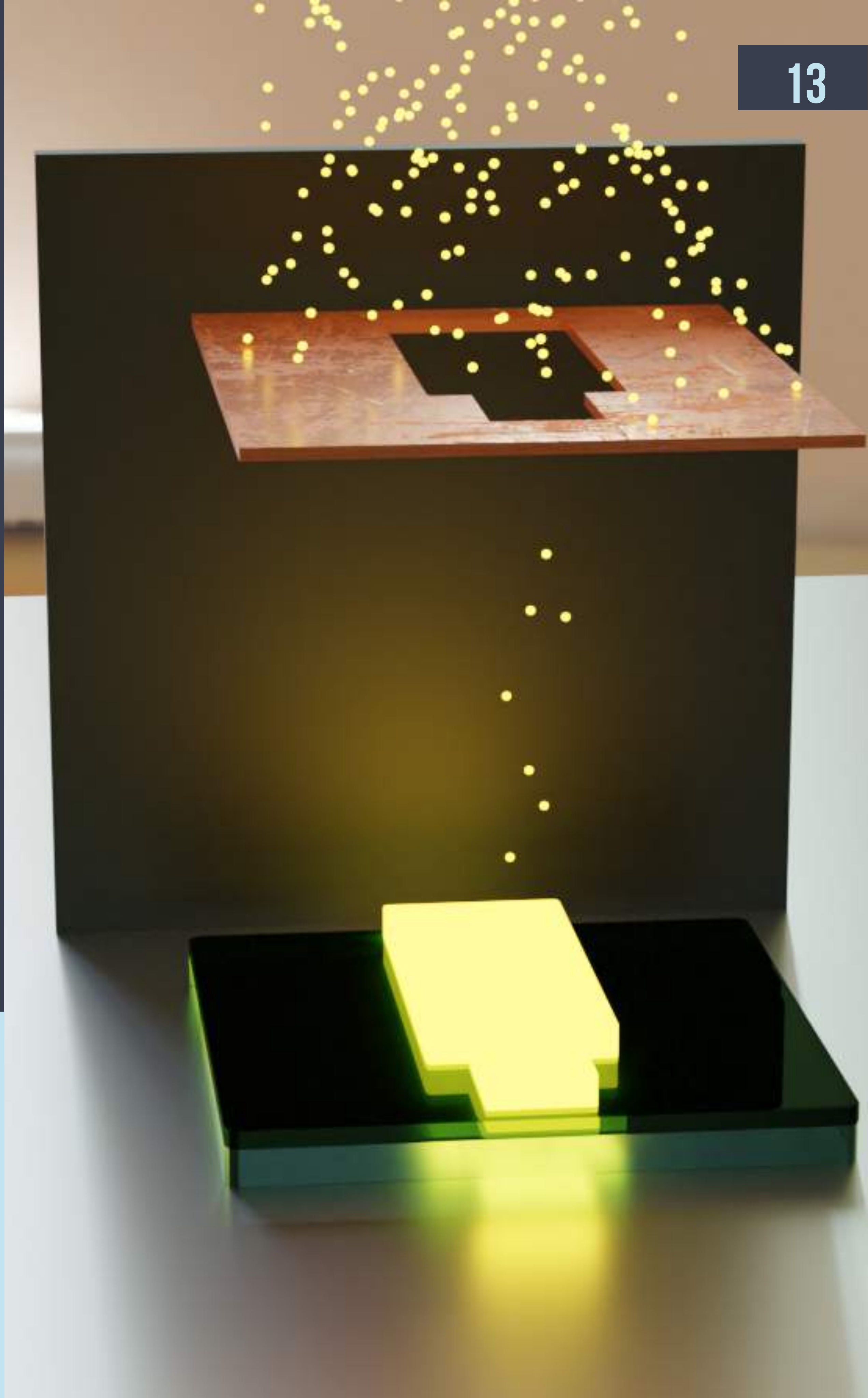
Thermal model of an electromagnetic radiation sensor based on a FI/S multilayered structure. Springs represent the thermal links, each one characterised by a thermal conductance G. When photons heat the absorber, they produce heat that flows through the whole system following these links. Eventually, the heat is released into the thermal bath.

CHAPTER 2

THE FI/S JUNCTION

The core of the SUPERTED sensor is a patterned multilayer structure, whose fabrication involves different material deposition and growth technologies.

In this section we go through the different steps required to build such structures, and the strategies used in the project for controlling the quality and achieving the best possible results.

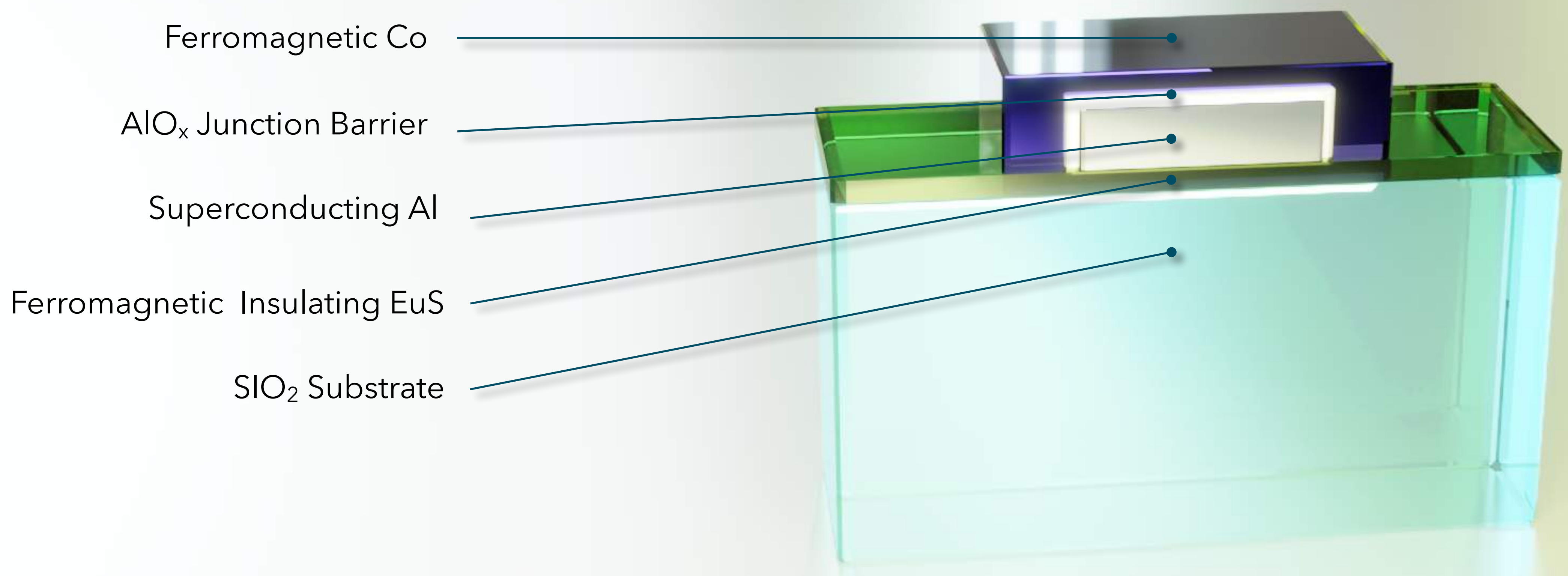


The development of the process required for the fabrication of the FI/S patterned structures was carried out in Work Package 1 of the SUPERTED project and mainly took place at Centro de Fisica de Materiales in Donostia-San Sebastian (Spain).

This research was led by:

• Celia Rogero (celia.rogero@ehu.eus)

•Max Ilyn (maxim.ilin@ehu.eus)



STRUCTURE OF THE FI/S JUNCTION

The most fundamental part of the SUPERTED sensor is the inner structure where the arrangement of several materials enables the Giant Thermoelectric Effect. Magnetic semiconductor Europium Sulfide (EuS) and Superconducting Aluminium (Al) create a spin-split Density of States (DOS) via exchange coupling. The strong electron-hole asymmetry in each one of the spin components can be used to generate a thermoelectric signal when connecting the superconductor to another electrode via a spin filtering junction. In this case, this corresponds to a thin layer of Aluminium Oxide (AlO_x), and a ferromagnetic (metallic) Co electrode.

It is worth stressing that the construction of the device still requires integration of additional parts, in particular radiation absorbers and additional electrodes, as shown in Chapter 3.

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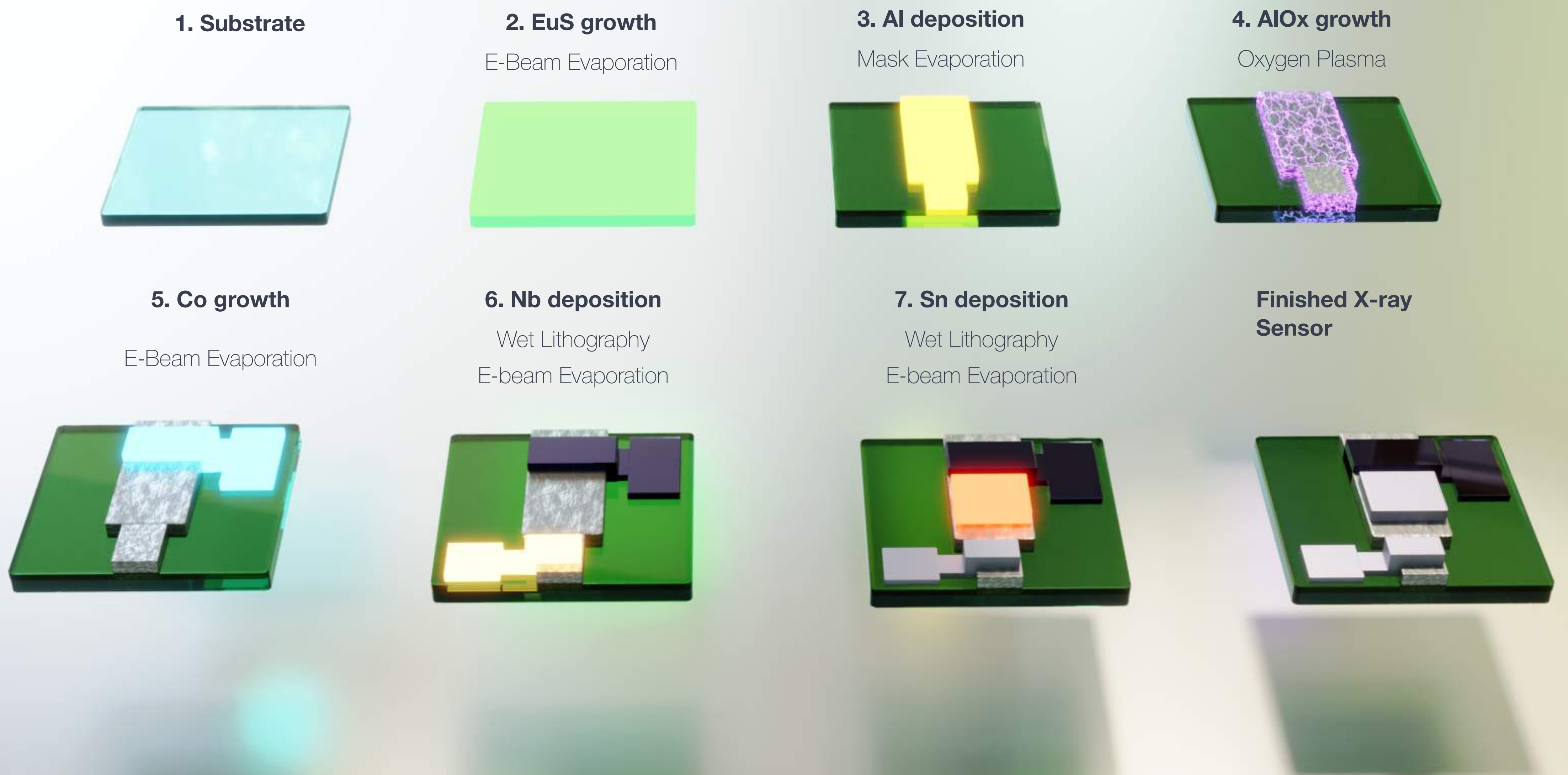
The information provided here focuses on the most direct way of fabricating the junction, but getting there requires a fairly large effort of trials and improvements. We encourage the reader to follow these discussions in Deliverable D1.1: *"Co/Al2O3/Al/EuS films" and* Deliverable D1.2: *"Spin filtering tunnel barriers".*

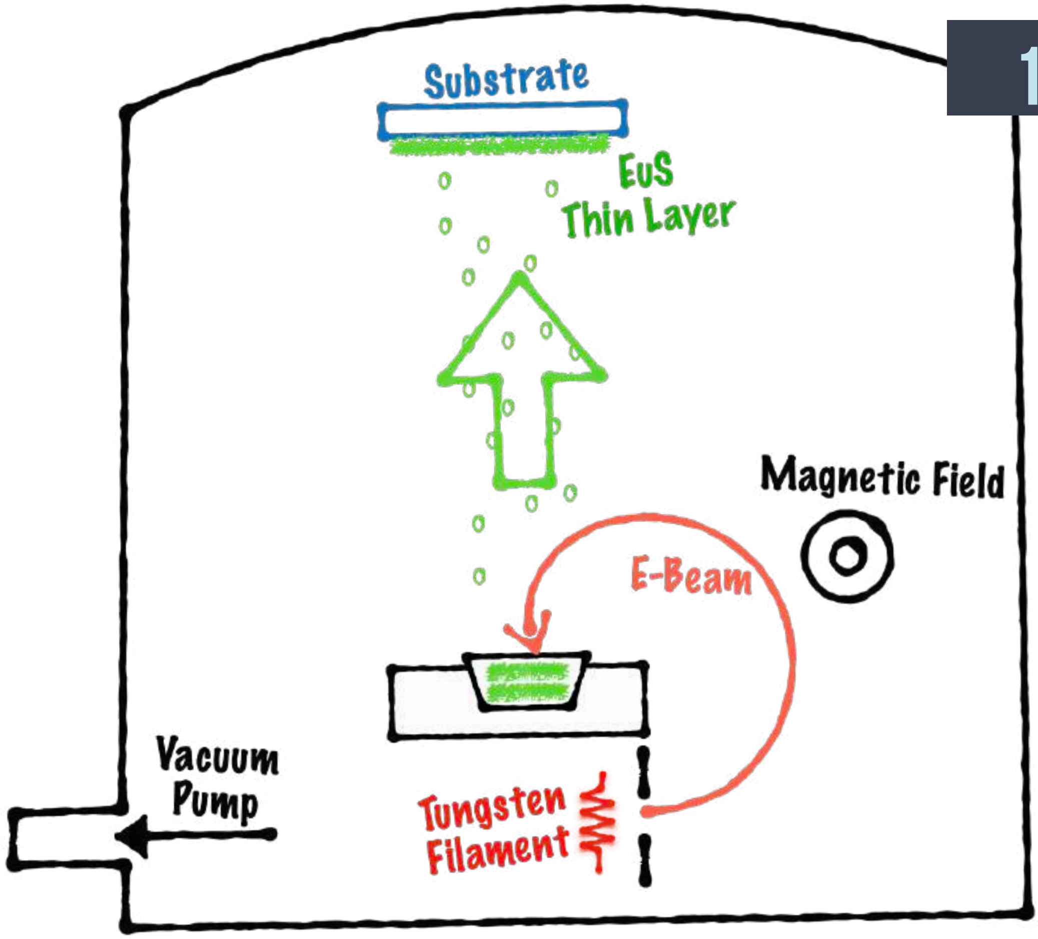


FABRICATION ENVIRONMENT AND PROCESS

The construction of the layered structure is a demanding task, because of several reasons. On the one hand, the exchange interaction which leads to the spin-splitting is very sensitive to the presence of contamination, such as the appearance of AlO_x, Eu_3O_4 or other EuS phases. On the other hand, the patterned materials have geometrical features and sizes which require accuracy in the range of tens of µm.

The best way of avoiding contamination is to carry out preparation of the multilayered structure in an Ultra High Vacuum (UHV) environment (with a base pressure better than 5 x 10^{-10} mbar). All different material deposition steps must be carried out sequentially in one single vacuum cycle, in order to avoid exposing the device to the pollutants of the atmosphere. The challenge is therefore to implement deposition techniques which 1) are compatible with the temperatures required to deposit each material, 2) are compatible with UHV environment and 3) allow for a relatively high resolution.

We now explore the processes used in SUPERTED for building the sensors.



THERMAL EVAPORATION: GROWTH OF EuS LAYER

In thermal evaporation processes, a source material is heated, and the the resulting vapour is directed towards a substrate, where it condenses again forming a thin-film layer. It is a form of Physical Deposition (that is, the deposited material does not necessarily create a chemical bond with the substrate).

The whole process takes place in vacuum environment, because of several reasons: 1) under such conditions the vapour pressure required to raise a vapour cloud is lower (i.e. the melting temperature of materials is lower than at atmospheric pressure) and 2) evaporated particles travel directly towards the deposition substrate allowing for a most accurate control of the deposition homogeneity and avoiding contamination due to collision of evaporated molecules with other molecules present in the atmosphere.

E-Beam evaporation is a particular type of thermal evaporation, in which the source material is bombarded with an electron beam emitted from a charged tungsten filament. This allows for a direct transfer of energy from the electron beam, allowing reaching higher and more localised temperatures than with other evaporation techniques. In practice, this means that it can be used for depositing materials with high melting points. The technique allows for high deposition rates (typically 0.1 nm to 100 nm per minute), which results in high-density film coatings with increased adhesion to the substrate.

The EuS layer is directly deposited on top of a clean substrate, and does not require any patterning (i.e., covers the whole surface of the substrate homogeneously). The source material which is evaporated is directly EuS (as opposed to using separated Eu and S sources).

The thickness of the EuS layer is one of the parameters that has an impact in the performance of the transducer. It has an influence over the exchange field that causes the spin splitting in the superconducting layer of the transducer. Too thick layers will suppress the superconducting state. Typical thickness of this layer in our sensing devices are in the range of 8-10 nm.

An essential characteristic of the EuS layer is that it must be fully stoichiometric (1:1 ration of Eu:S), which is only possible if deposited Eu ions are in the Eu^{2+} state with spin moment S=7/2 and zero orbital moment. Appearance of Eu^{3+} ions lead to the appearance of other chemically active phases, such as Eu_2S_3 or Eu_3S_4. The way to avoid this problem is to keep the target area at a low temperature. A simple way of achieving this is to pre-cool the substrate using liquid nitrogen. Keeping the target surface at a low temperature has the additional advantage of leading to a low roughness of the deposited EuS layer, which is required for allowing the correct deposition of the next layer (superconducting Al).

A RMS roughness of 0.75 nm in the EuS layer is deemed sufficient for allowing the growth of good quality Al.

CHARACTERISATION METHODS

The set up and optimisation of a deposition process requires applying characterisation techniques that could provide information about the resulting structures.

For the purpose of determining the quality of the EuS layer, the following techniques have been used in the SUPERTED project:

- **X-ray Photoelectron Spectroscopy (XPS)** can be used to determine the chemical composition of the thin film layer, and in particular to identify the state of the Eu ions. This allows tuning the growth temperature.
- **Atomic Force Microscopy (AFM)** can be used to characterise the topology of the surface of the EuS layer, as its roughness has a strong influence over the quality of the deposition of the Al layer.

These techniques can be applied in the same UHV environment where the EuS evaporation takes place.

Concerning the impact of the thickness of the EuS layer in the system, magnetisation measurements can be carried out in order to have an idea of the strength of the interaction. Saturation m (i.e., the value of an applied external magnetic field H at which the magnetisation of the material does not increase significantly any further) evolves linearly with the thickness of the EuS layer.

The use of these techniques is described in detail in Deliverable 1.1.

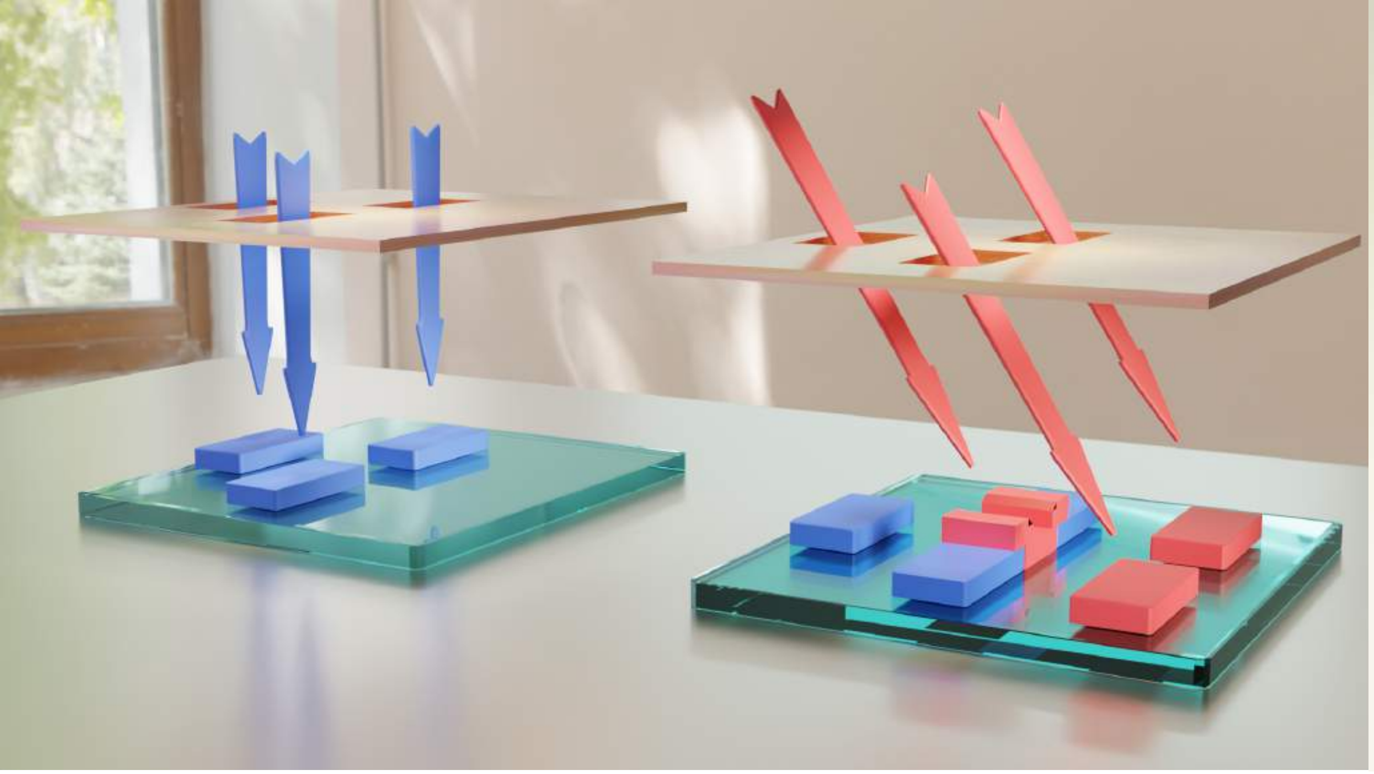
SHADOW MASK DEPOSITION: AL AND CO LAYERS

Once the Ferromagnetic Insulator has been deposited (covering the full surface of the substrate), the remaining layers have to be given a three-dimensional patterned structure, which allows guiding the flow of carriers towards the electrical contacts for integration of the device with the read-out electronics that extract the output signal.

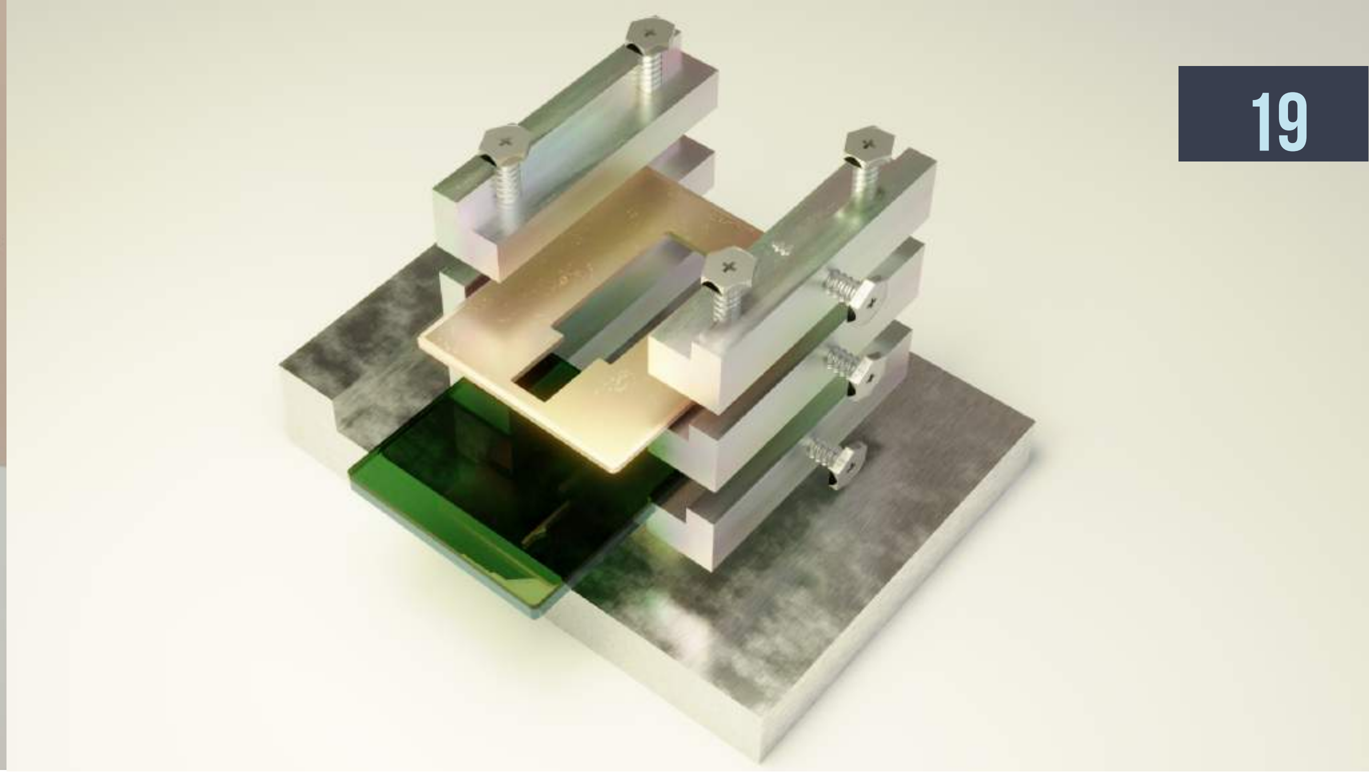
These layers consist of metals (Aluminium, Al and Cobalt, Co) which constitute the superconducting and ferromagnetic, respectively, layers of the junction. They are separated by a thin layer of Aluminium Oxide (AlO_x) which is generated through plasma oxidation as it discussed below.

The deposition process used for building these layers can also be thermal evaporation, but it has to be further adapted so that a geometrical pattern can be transferred. Several techniques can be employed for this (such as photo-lithography) but one specially simple, and compatible with the process in the UHV environment has been applied in the construction of the SUPERTED sensors.

The shadow mask technique takes advantage of the fact that in the UHV environment the thermal evaporation process can be optimised so that the evaporated molecules travel in an almost straight line until they reach the target surface. A very straightforward way of transferring a pattern is then to interpose a mask, consisting of a sheet of material where a



Shadow mask deposition using different sources: The same mask can be applied sequentially in order to deposit several materials. Sources must be oriented differently.



Implementation of shadow mask deposition in a UHV environment. A special holder is used to keep in place the target substrate and one or more masks.

series of apertures let the evaporated beam reach the target only in specific locations of the target substrate.

In its most basic set up, shadow masks are prepared as exact negatives of the pattern which has to be transferred. In this case the mask must be placed in a plane perpendicular to the direction of the beam of evaporated material. If this angle between the mask and the beam is modified, the shape and the location of the deposited pattern are different. This is an interesting feature which allows using the same mask for depositing different layers, each one with a different geometry (if the layers are made of different materials, several sources must be integrated in the evaporation chamber at the same time).

Another advantage of the shadow mask is the relatively simple hardware required. It basically consists of a modular holder which hosts the target substrate at the bottom, and one or more shadow masks on top. The main parameter to adjust is the distance between the mask and the layer, which largely depends on the complexity of the process (if mask is going to be used for deposition at different angles, several materials, etc). There is, however, a maximum distance limit because

SUPERTED Detailed information about the design of the shadow mask equipment used in the SUPERTED project can be found in section 3.1 of Deliverable D1.1. *"Co/Al2O3/Al/EuS films"* collisions with the edges of the mask change the direction of part of the evaporated material. If the time of flight to the substrate after this collision is large, then the edges of the patterned structure are not sharp. This is ultimately the main limitation of the technique, which makes it unsuitable for patterns with very small dimensional features (this is not the case of current designs of SUPERTED sensors, where features are in the range of hundreds of microns).

The dimensional parameters of the Al layer play an important role in several aspects of the device:

- *The thickness of the Al layer affects the strength of the exchange field established due to the EuS layer. Strength of the field is inversely proportional to the Al thickness, as long as it does not become longer than the superconducting coherence length (which is typically of the order of 10^{-5} cm). Typical values of Al thickness fall within the 10-20 nm range.*

- *The optimal quantum efficiency of the transducer is attained when the normal resistance of the Al layer is matched with the impedance of the absorber/antenna. For a given thickness, the width and length of the layer can be modified as long as the ratio remains constant. In the SUPERTED project, both dimensions were kept at the same nominal value of 200 μm.*

In what concerns the Ferromagnetic (Co) layer, in principle the relevant geometrical features are those determining the area of the junction. Since the superconducting and ferromagnetic layers normally cross in orthogonal directions, the width of the Co layer is the feature that determines the amount of overlapping between both layers.

CHARACTERISATION METHODS

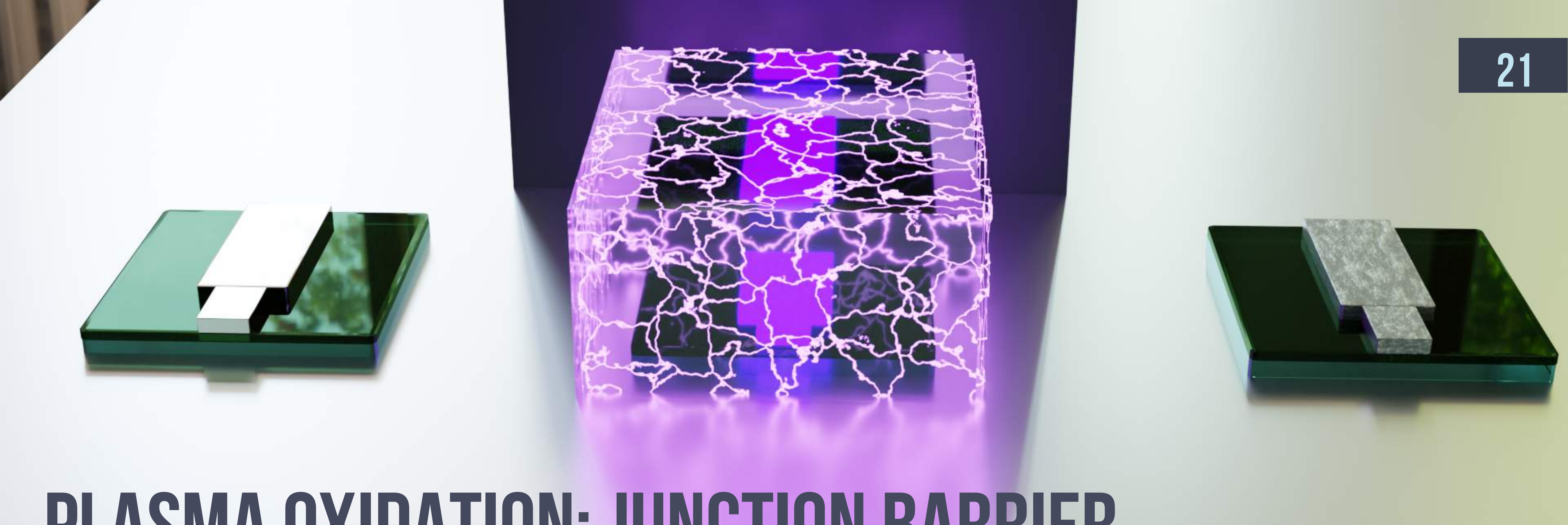
In principle, the lateral dimensions (length and width) of a layer deposited using shadow mask technique, if properly set up, must not vary significantly from nominal specifications (defined by the mask). In any case, given the dimension in the range of many μm, optical microscopy, or atomic force microscopy (AFM) can be used to determine the dimensions.

The thickness of the layers can be monitored on real time during the deposition process using a quartz crystal sensor.

Quality of the structure is ultimately determined by its electrical behaviour. Resistance measured across the junction, using the 4-point probes method for better accuracy, should allow determining the resistivity. These values can be compared with, and should be close to, those of pure Al, although thin film resistivity is typically higher than that of thick bulk samples, because the film thickness often places an upper bound for the elastic mean free path and thereby affects resistivity.

Details about this type of measurements can be found in Section 3.2 of Deliverable 1.1.

The behaviour of the tunnel junction is best characterised by measuring the differential conductance, which can be either directly measured with the lock-in technique or calculated as a numerical derivative of the I-V curves measured across the junction using a 4-point probe set up. A detailed discussion about such characterisation can be found in Deliverables *1.2 - "Spin Filtering Tunnel Barriers"* and 1.3 - *"DOS measurements"*, as well as in the publication *"Revealing the magnetic proximity effect in EuS/Al bilayers through superconducting tunneling spectroscopy".* Phys. Rev. Mat., 1, 054402 (2017). DOI: doi:10.1103/physrevmaterials.1.054402



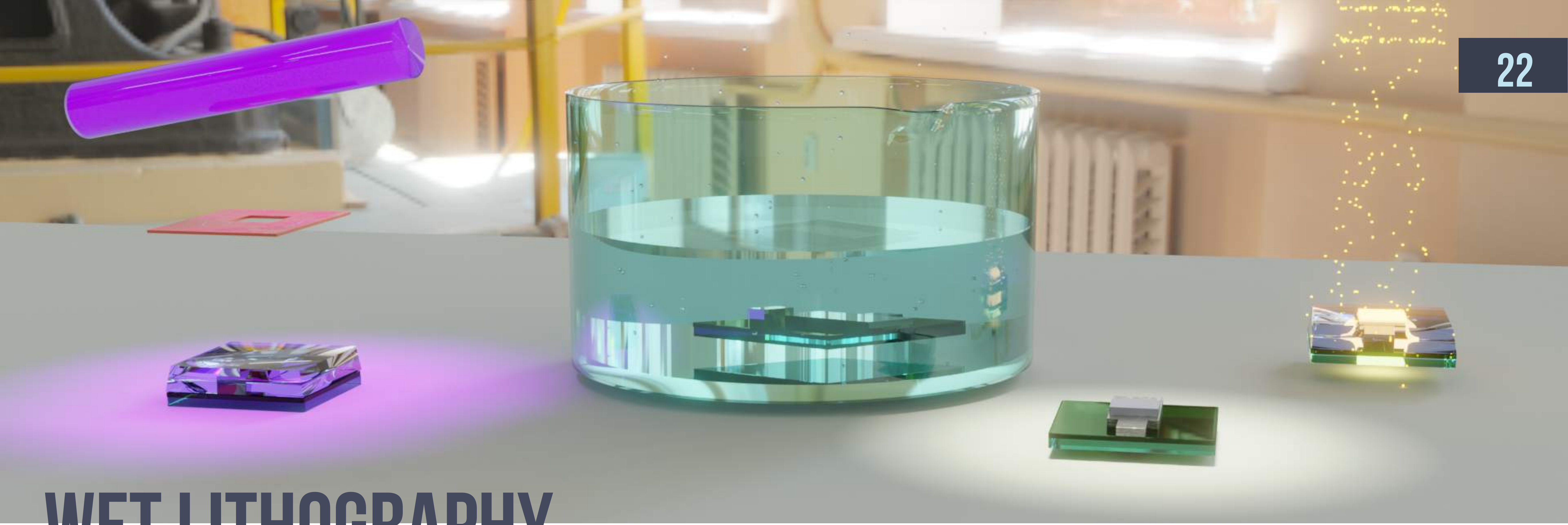
PLASMA OXIDATION: JUNCTION BARRIER

The thin layer of Aluminium Oxide (AlO_x) located between the Superconducting Al layer and the Ferromagnetic Co layer acts as a tunnel junction. It has a strong influence on the operation of the junction. The junction can be characterised through its spin-averaged normal-state conductance G_T. This value depends linearly on the area of the junction and exponentially on the thickness of the tunnel barrier, so the latter parameter can be used as a way of achieving a precise control over G_T. Typical thicknesses of the AlO_x layer are in the range of 2-3 nm.

Growth of the AlO_x layer is done by means of direct oxidation of the Al layer, using an oxygen plasma set up. This technique is compatible with the UHV environment, and with the mask evaporation technique. Oxidation must be applied between the deposition of the Al and Co layers.

The plasma oxidation technique is commercially available, and consists of a radio frequency current which is fed into a coil that surrounds a discharge tube, so that the corresponding field is inductively coupled and plasma is generated from the gas molecules inside the plasma discharge tube.

The AlO layer grown through this procedure is actually a coating that covers all the surfaces of the Al layer. Such a layer has to be removed from the places where ohmic contacts are required once the procedure inside the UHV environment has finished.



WET LITHOGRAPHY

The removal of the AlOx layer in specific places requires the use of sophisticated masking techniques, which require extracting the device out of the UHV environment. It is worth stressing that there are alternatives to the use of selective masking, such as Argon plasma sputtering, which have indeed been tested during the SUPERTED project. It has, however, been deemed that such a technique is too aggressive and produces irreversible damage in the junction.

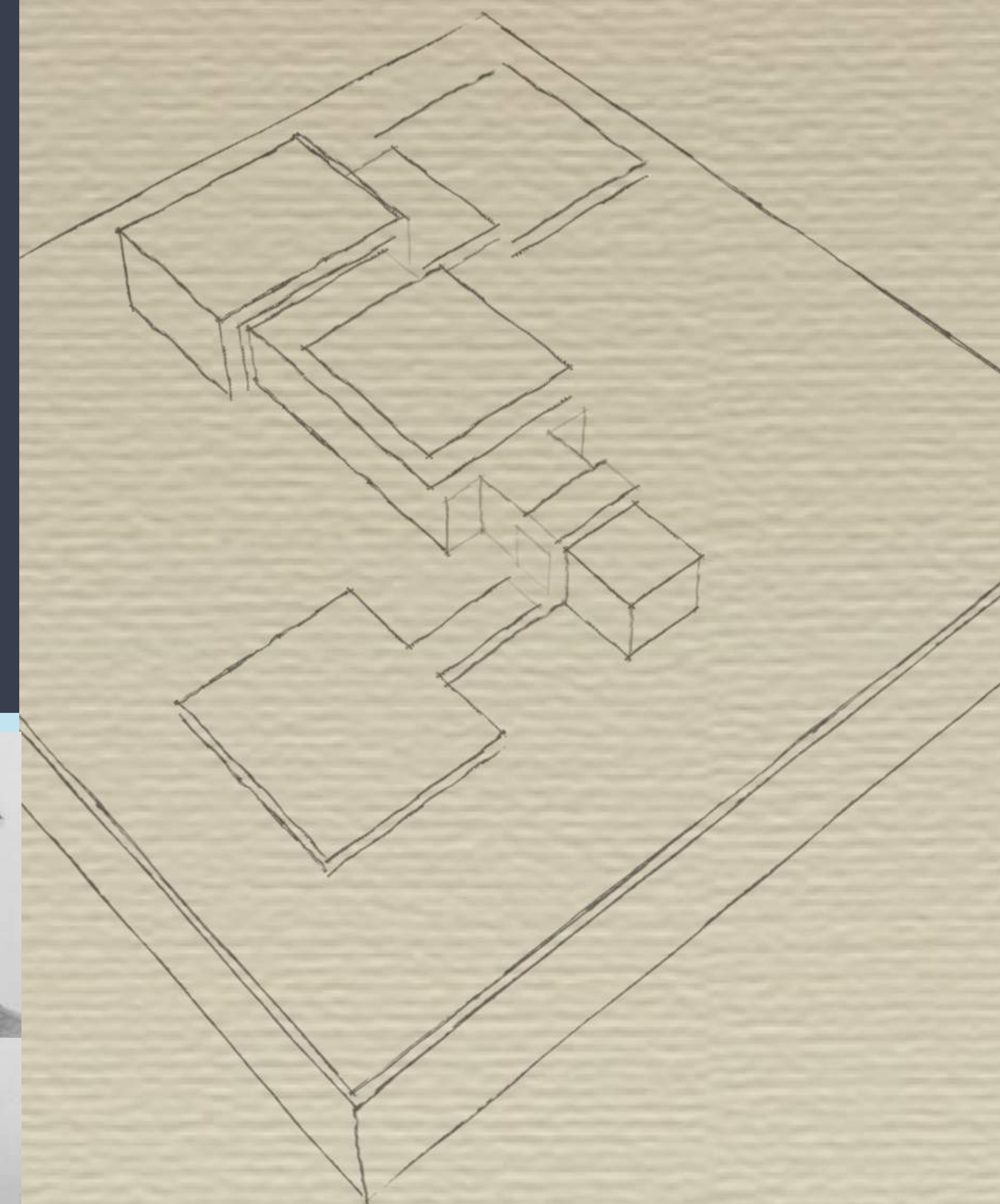
Wet etching techniques are rather work intensive, but allow for highly controllable and reproducible results. The concept is to create a protective coating in localised areas of the device, and then applying an etching process that will chemically react with all surfaces which are not protected by the coating. The coating consists of a polymer substance which can be hardened using Ultra-Violet (UV) light. A negative of the pattern to be transferred is interposed between the UV source and the device, so that the created shadow leaves zones with unhardened polymer, which is then washed away. The chemical edging solution has to be adapted to the materials. In this case, a solution of Tetramethylammonium hydroxide (TMAH) in pure water at 1.25% concentration can be used for the removal of the AlOx layer. After extraction from the chemical bath and washing, the whole polymer coating is removed, leaving the device ready for further processing.

Deposition of the remaining layers can be done using evaporation or sputtering (in the case of Nb electrode, for instance, high melting point makes it more suitable to use sputtering).

CHAPTER 3
SENSOR STRUCTURES

Sensors based on the Giant Thermoelectric Effect must integrate the FI/S junction that enables this physical phenomenon. This core must be coupled smoothly to other elements that allow the sensor to interact with the environment (absorbers or antennas) and to send the output away (read-out).

The way all these elements are integrated ultimately determines the device performance. Several parameters, regarding the type of materials and the geometrical features, can be used for optimising this process, as we show in this chapter.



The development and optimisation of the sensors in the SUPERTED project was carrier out in Work Packages 2 and 3, and took place at the University of Jyväskylä (Finland), Institut Néel (France) and Scuola Normale Superiore - NEST (Italy). This work was led by:

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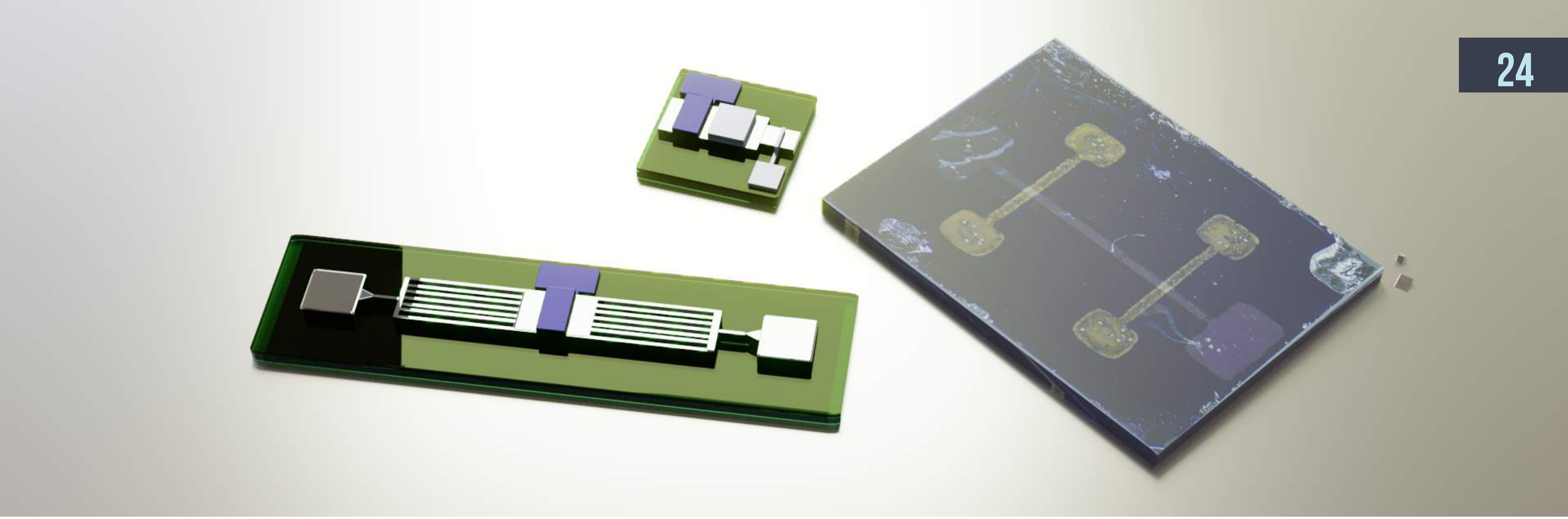
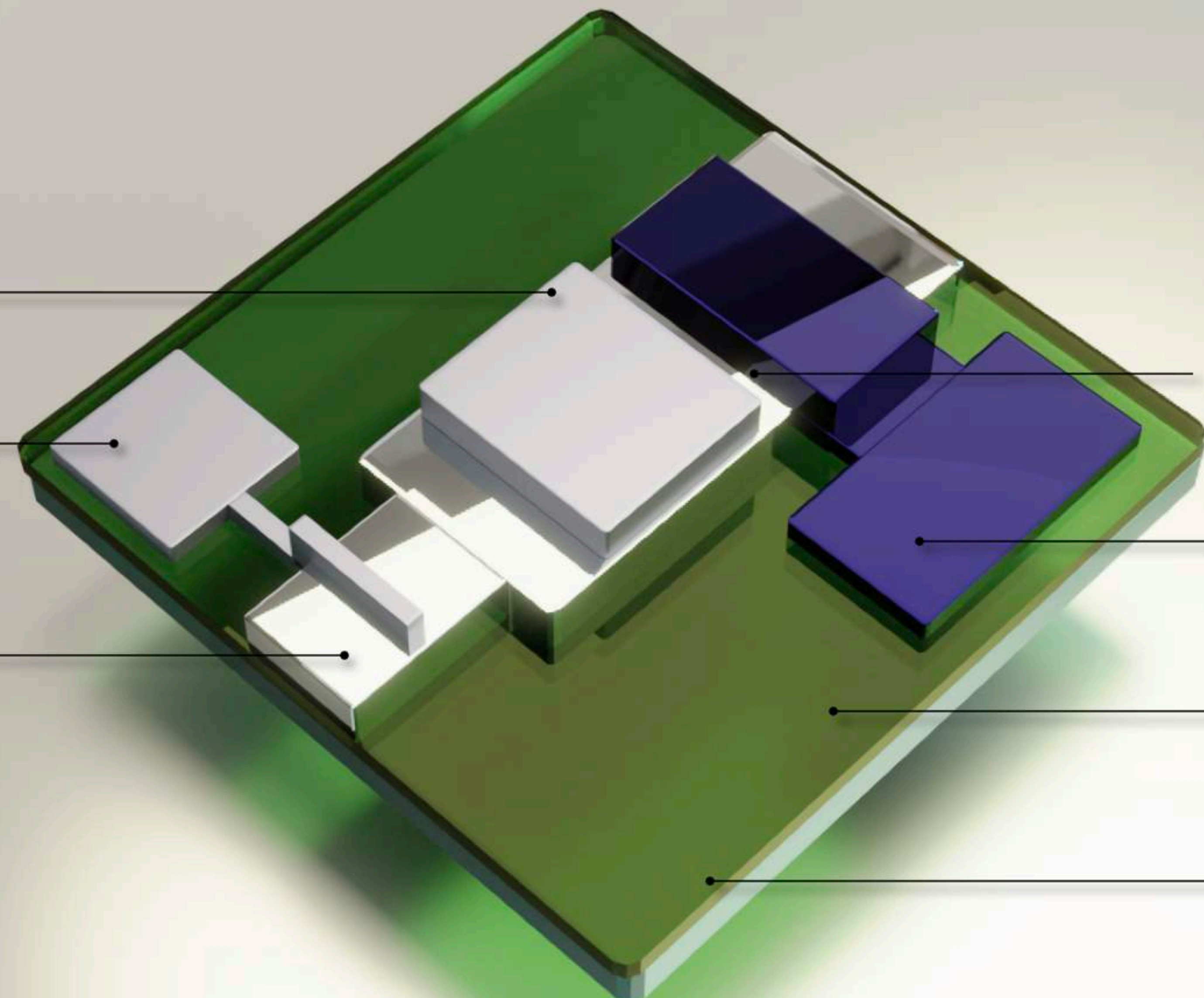


ADAPTING SENSORS TO APPLICATIONS

The use of high performance sensors unavoidably requires an adaptation an optimisation of the devices in view of the particular requirements of the application. An important part of this process implies tuning a set of parameters, which mainly relate to the type of materials used and the geometrical dimensions of the different elements of the device.

Given the many technical challenges that had to be overcome to reach the first realisation of the devices that took place in SUPERTED, we adopted the strategy of using samples with a generic geometry, each one of them containing two FI/S junctions, that made it relatively easy to carry out the different tests. Hundreds of these samples were prepared during the project, exploring different parameter combinations. The best samples where then employed in the fabrication the first sensor prototypes. This has allowed the technology to evolve one step further. We have now a good understanding about the structures of the optimal sensors, and how they can be engineered for optimal performance in a specific application. These are the ones that are presented here.

As mentioned above, thermal sensors can be configured to work in two different regimes (as micro-calorimeter or as bolometer). Since there are significant difference between them, we discuss them separately.



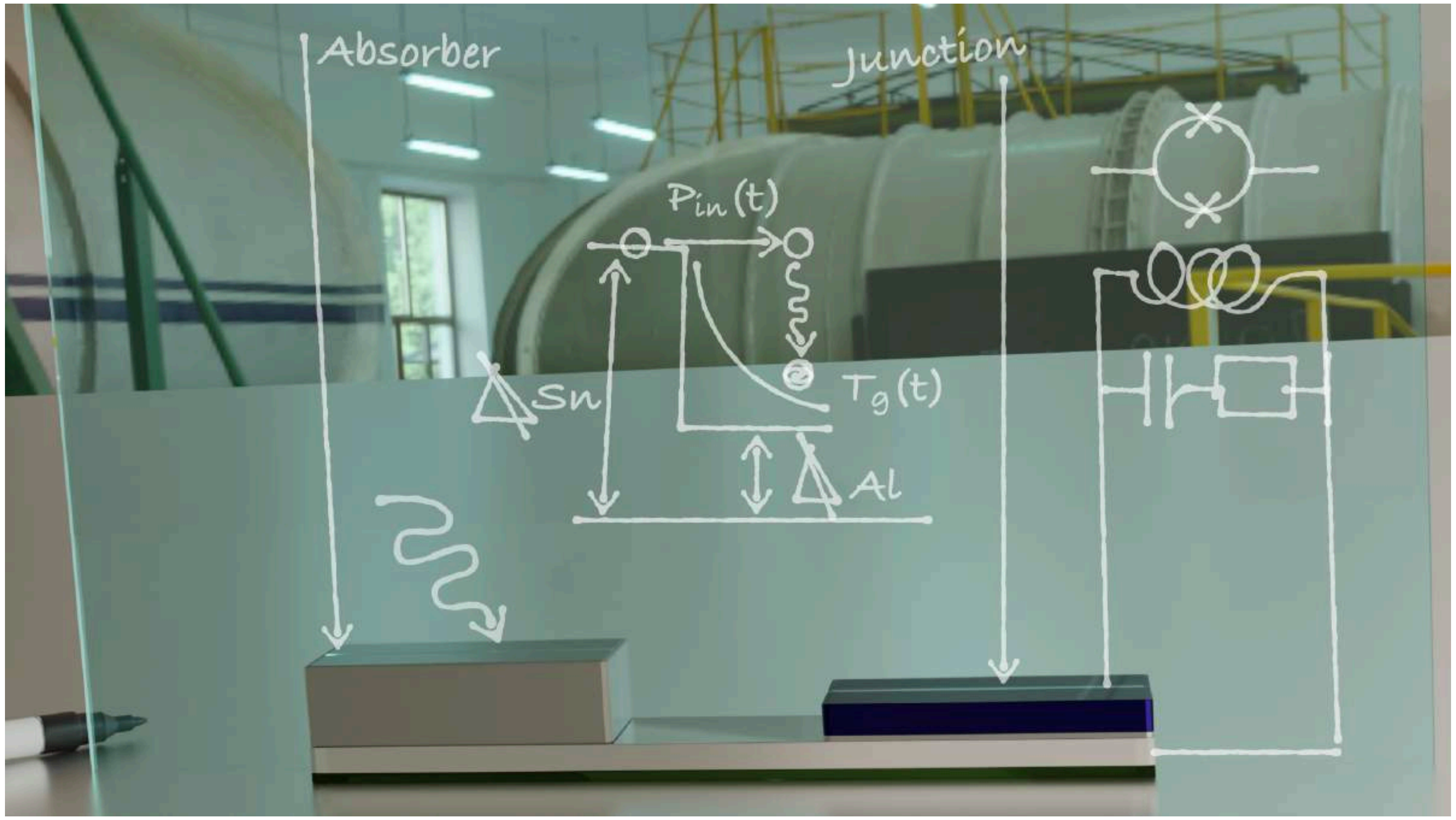
X-RAY CALORIMETER

The X-ray sensor is basically composed of an absorber, which converts energy of incoming photons into thermal signals, and a transducer which converts thermal signals into electrical signals and drives them towards the read-out electronics (through the electrodes of the device). The resulting electrical signal is proportional to the absorbed energy. The thermal signal is finally drained out through the heat sink (substrate), and the sensor will return to the equilibrium state, and be ready for another sensing cycle.

Tin (Sn) has been chosen as the material for the absorber. It is a superconductor at the operating temperature of the device, and offers a good compromise between high stopping power (high-Z) and low specific heat capacity.

When a photon hits the Sn layer it creates a cloud of quasiparticles resulting from the breaking of Cooper pairs. These quasiparticles are transferred to the (also superconducting) Aluminium (Al) layer, which is already part of the transducer. The superconducting gap of Sn is higher than that of Al, and therefore prevents heat from leaking outside the absorber.

The Aluminium layer is placed on top of another layer of Europium Sulphide (EuS) which behaves as the ferromagnetic insulator, inducing an exchange field which causes spin splitting in the superconducting density of states of Al. The electron-hole asymmetry leads to the giant thermoelectric response in a junction formed by the Al layer, a tunnel barrier, made of Aluminium Oxide (AlO_x) and a ferromagnetic material, which has been chosen to be Cobalt (Co). The electrical current generated by the aforementioned thermal effect, is driven by the (electronic) temperature difference between the Al superconducting layer and the Co ferromagnetic layer (that of Al is higher since it generally has a lower heat conductance to the phonons).



Radiation sensor based on a FI/S transducer. When a photon hits the superconducting absorber, it generates a population of quasiparticles which is proportional to the released energy. These quasiparticles travel through the superconducting layer of the transducer towards the junction.

The extraction of the electrical signal can be done by establishing an electrical read-out circuit through two electrodes. The Co layer itself can be used as one of the electrodes, while the remaining one can be fabricated depositing a Niobium layer in direct contact with the Al one. Care should be taken to make sure that the size of the Nb element is small enough, in order to minimise heat leakage.

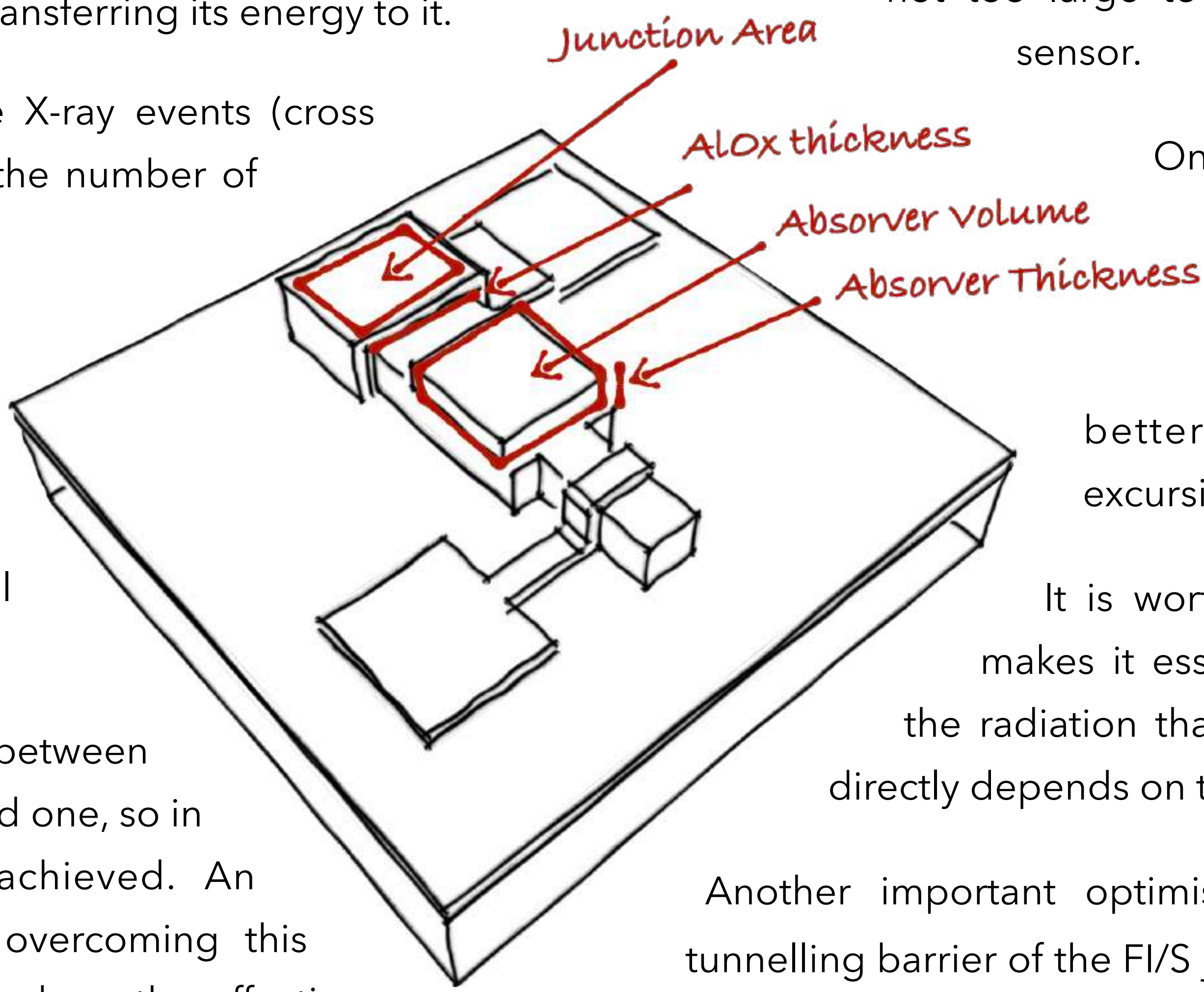
In the first approximation, the read-out circuit connects a capacitance C and an inductance L in parallel. Taking into account that the whole sensor must be placed in cryogenic environment during normal operation, extraction of the signal is not so straightforward, but can be done using well known techniques based on the use of SQUID amplifier readouts. This topic is covered in Chapter 4.

The geometrical dimensions of the different layers play a role in the final performance of the device. This is particularly true in the case of the absorber layer, where several parameters have an impact in the efficiency of the collection of the X-rays:

- **Thickness** determines the absorption efficiency of the incoming X-rays (also known as stopping power). In principle, thicker layers are better for this task, as each photon interacts through a greater length with the material, transferring its energy to it.
- **Area** determines the rate of the X-ray events (cross section). A larger area increases the number of absorbed photons.
- **Volume** determines the heat capacitance, which dictates the peak signal and the ultimate energy resolution. Larger volume reduces the signal level and degrades the resolution.

There is an obvious antagonism between the first two parameters and the third one, so in practice a trade off must be achieved. An alternative strategy which allows overcoming this issue is to use arrays of detectors, where the effective cross section equals the sum of the areas of the individual absorbers.

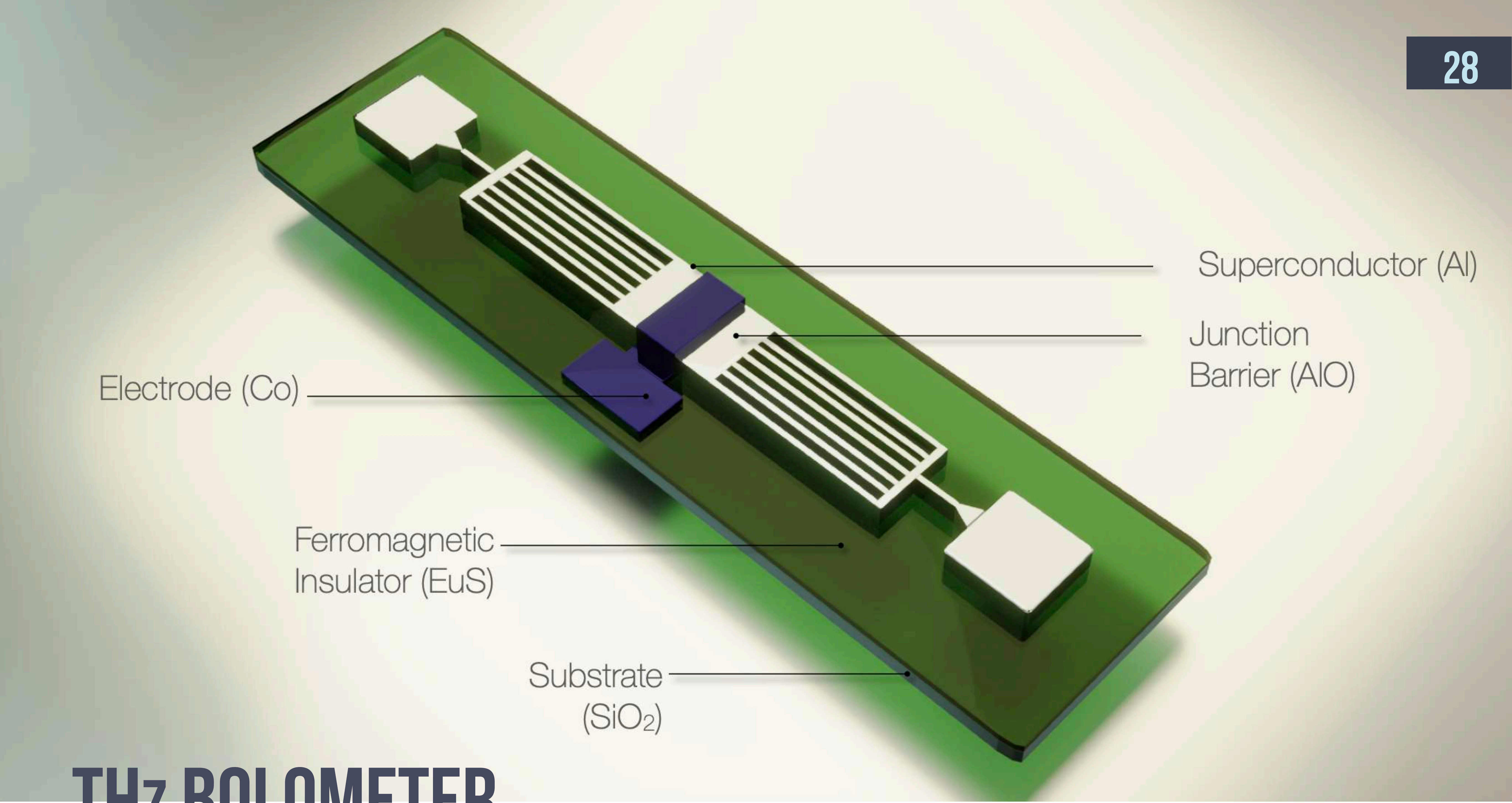
The optimisation of absorber volume should be based on the application. The trade-off in design of the absorber volume is between the temperature excursion (dT) and the energy resolution of the detector (dE). On the one hand, the temperature excursion, defined as $dT = \frac{E_{particle}}{C_{absorber}}$, corresponds to the temperature change caused by the absorption of energy from an incoming photon ($E_{particle}$) and is determined by the heat capacitance ($C_{absorber}$). In principle, it is desirable that dT is large, but not too large to overcome the linear regime of the sensor.



On the other hand, the energy resolution dE is (roughly) proportional to $\sqrt{C_{absorber}}$. For high resolution applications, the smaller dE the better, but this degrades the energy excursion.

It is worth stressing that all this optimisation makes it essential to define the energy range of the radiation that has to be detected, which in turns directly depends on the desired application.

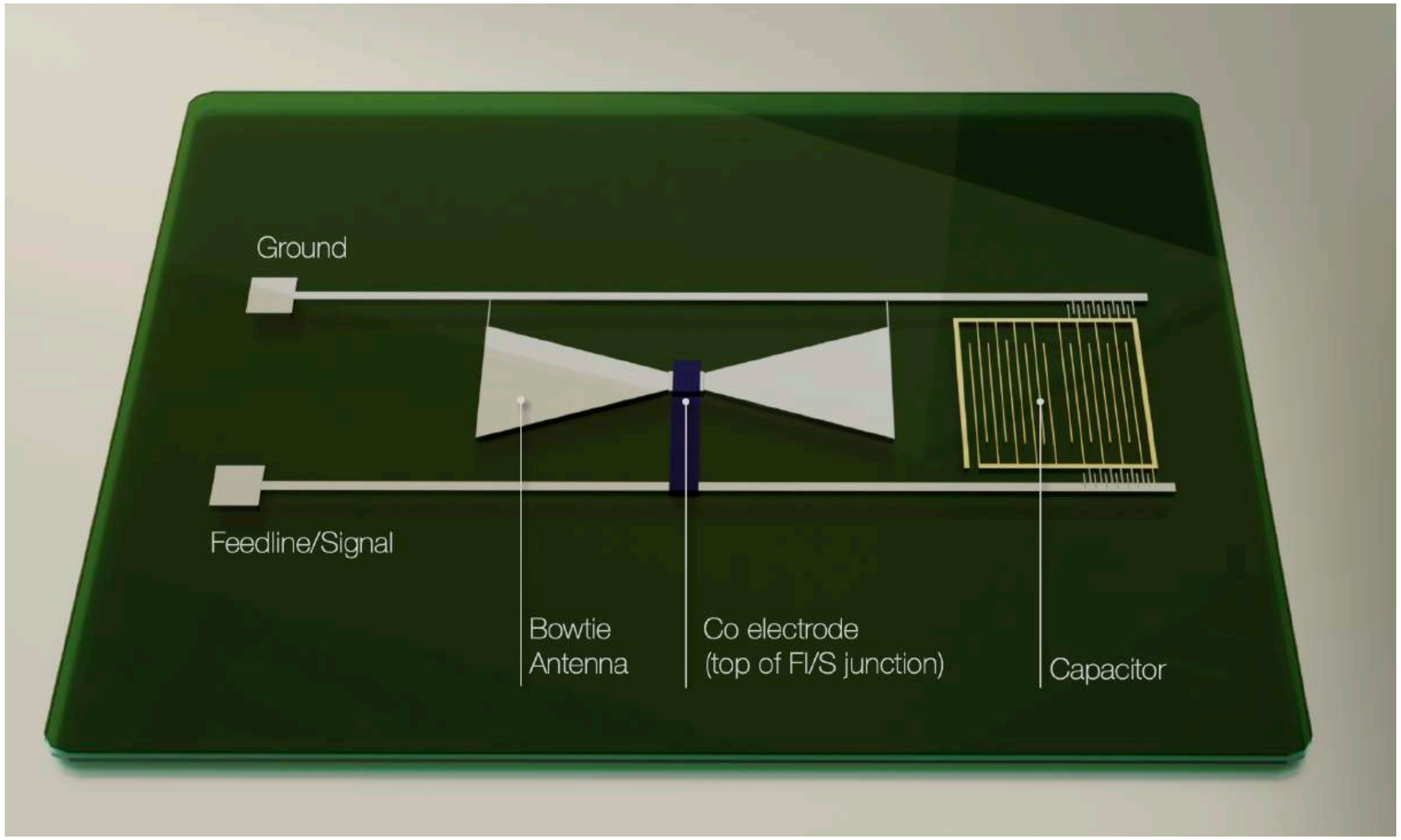
Another important optimisation parameter is related to the tunnelling barrier of the FI/S junction. The tunnelling resistance (R_T) has an influence in several aspects of the detector. Larger values of R_T reduce noise, but also reduce the signal level and lower the detector speed. The geometrical parameters with direct influence over R_T are the thickness of the AlO_x layer and the area of the whole junction.



THz BOLOMETER

In this configuration, optimised for detection in the THz/GHz range, the incoming radiation is coupled and guided towards the transducer using antennas made of superconducting material. A very straightforward way of building the device is to carve the antennas in the same Aluminium layer which makes part of the transducer. This simplifies the number of steps required for manufacturing the device, but has an impact in the overall performance, because the thermal diffusion between the antenna and the transducer are unhindered, causing thermal fluctuations that induce noise (typically this is reflected in the Noise-Equivalent Power, NEP, factor). A way to avoid this situation and improve the performance is to fabricate the antennas in a superconducting material with higher band gap than that of the transducer. This blocks the heat leaks, and limits the energy exchange between the antenna and the transducer to the electron-phonon coupling.

In principle any valid types/shape of antenna (meander, bowtie, dipole, etc) can be adapted to the sensor. Quantum efficiency can be optimised by adjusting the normal-state resistance of the Al layer with the impedance of the antenna. This can be done by cutting out material from the antenna, which has the effect of increasing average resistance towards the vacuum resistance.



Bolometer integrating Tiebow antennas for detection of radiation in the THz range, *A nearby capacitor is used to create a resonator. The feed line on top of the image is used to read the changes in amplitude and phase of a bias signal. The line between the sensor and the capacitor is used as ground.*

In what concerns the transducer, it is composed directly by the FI/S junction which has been described in chapter 2, that is, the Al superconducting layer is placed on top of a EuS layer, then the Al is oxidised to create a layer of AlO_x which acts as a tunnelling barrier between the Al and a layer of ferromagnetic Co. The latter can be used as an electrode. Unlike the case of the micro-calorimeter, there is no need of integrating an additional electrode, as the antenna can be connected to the ground for closing the electrical circuit.

An important aspect concerns integration into large arrays, because many applications in the THz/GHz range are related to imaging systems which use focal plane arrays for detection. The challenge in this case is to implement an efficient way of

reading the signals of each one of the individual sensors (pixels of the array). We propose to use a read-out scheme inspired on those already demonstrated for other types of superconducting sensors and, in particular, kinetic inductance detectors (KID). These sensors are able to detect the changes in the population of Cooper pairs and quasiparticles, which are modified by the energy brought in by the thermoelectric current (which causes breaking of some Cooper-pairs into quasiparticles). The change in quasiparticle population can be detected as a change in the kinetic inductance, by coupling the antenna-transducer system (which act as an inductor) to a capacitor, in order to form a high quality factor resonator. The change in kinetic inductance induces a change in the resonant frequency and in the quality factor. This can be measured by measuring the change in the amplitude and phase of the bias signal of the resonator, transmitted past the resonator through a feed line.

CHAPTER 4
INTEGRATION

The final step for having a working device is to couple it to the signal read-out circuits and to integrate it inside a proper working environment. As any superconducting sensor, the SUPERTED sensors will require deep cryogenic temperatures for operating.

We discuss these points in this chapter, and present the work done in the project for demonstrating the sensors in two emblematic applications: X-ray spectroscopy for the micro-calorimeter and THz astrophysics for the bolometer.

The design of the read-out system has been carried out in WP3.

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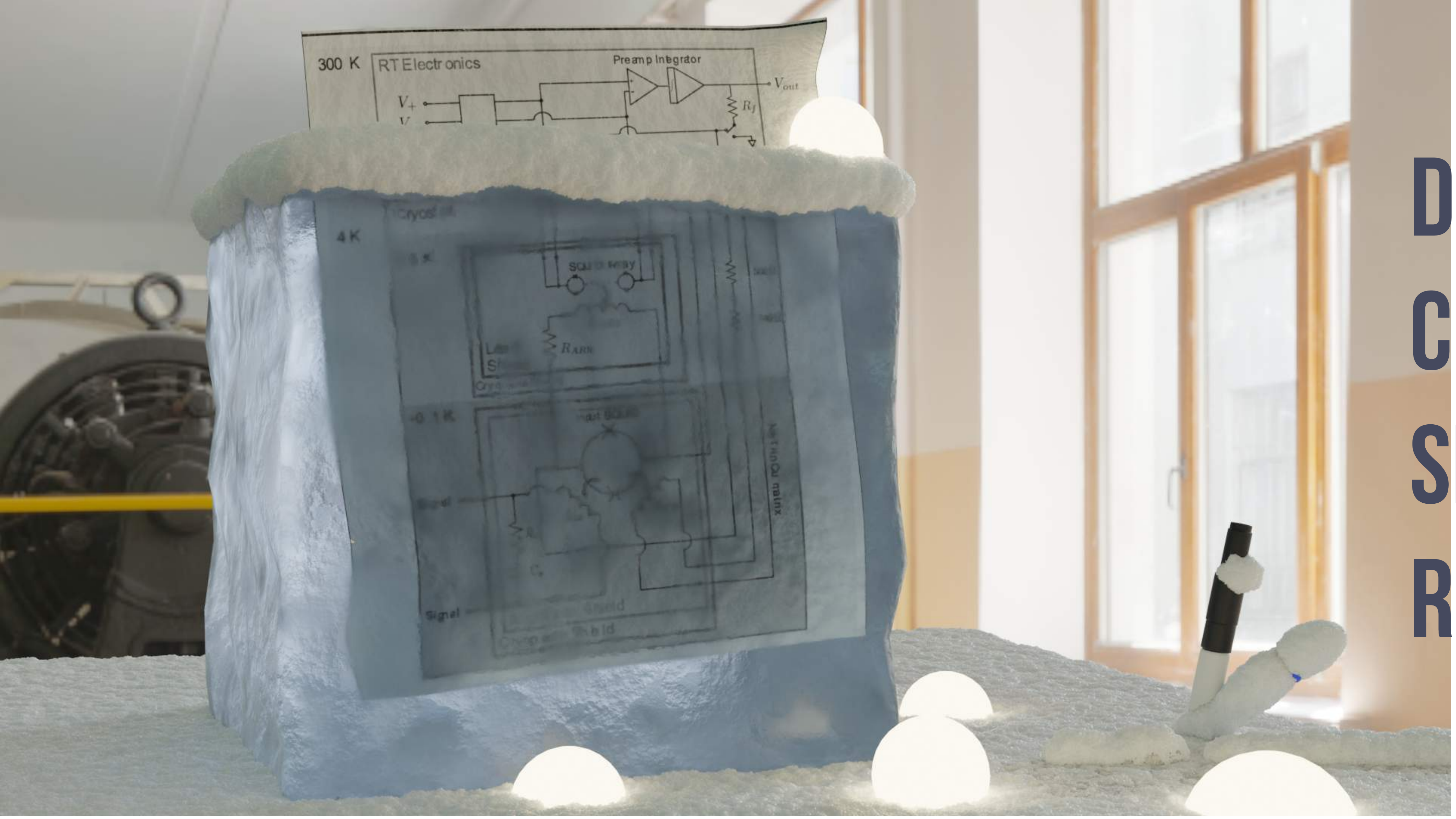
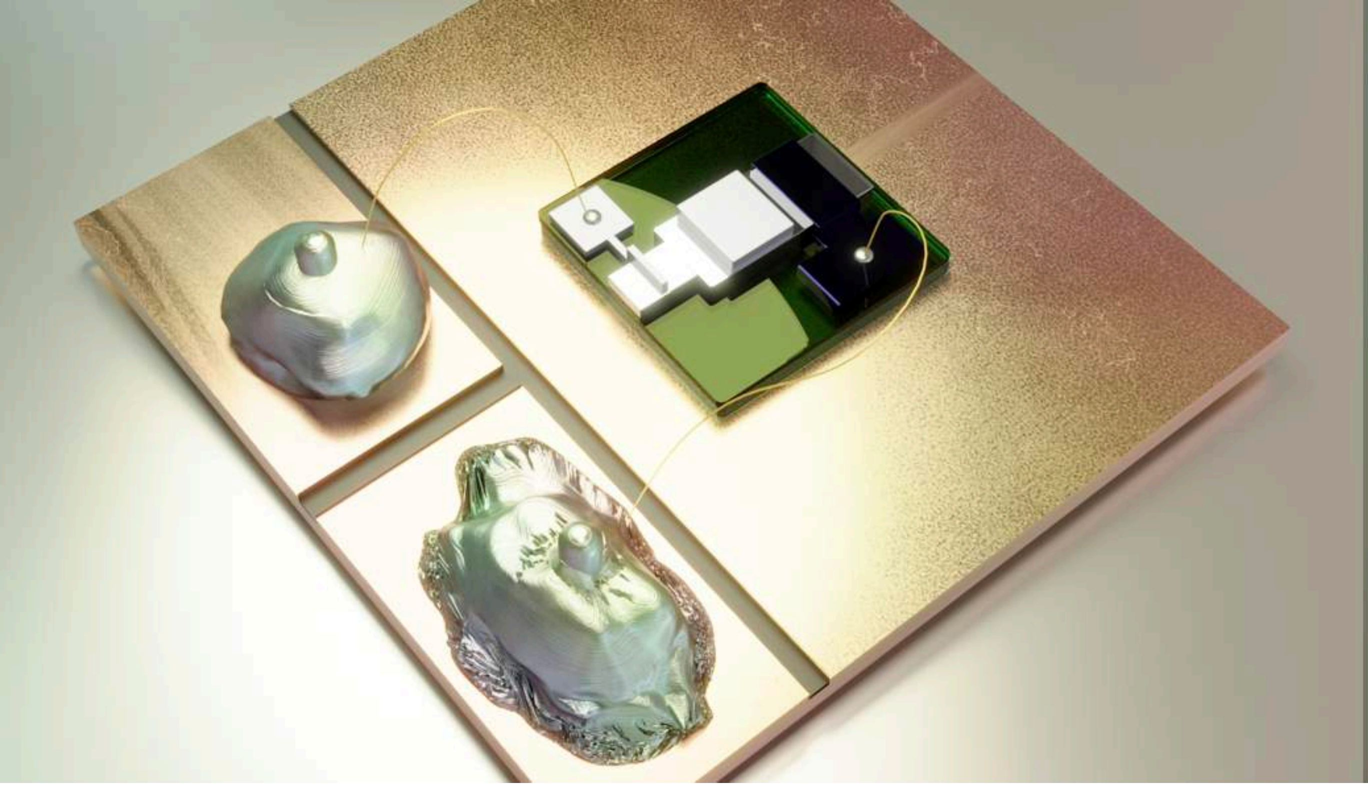
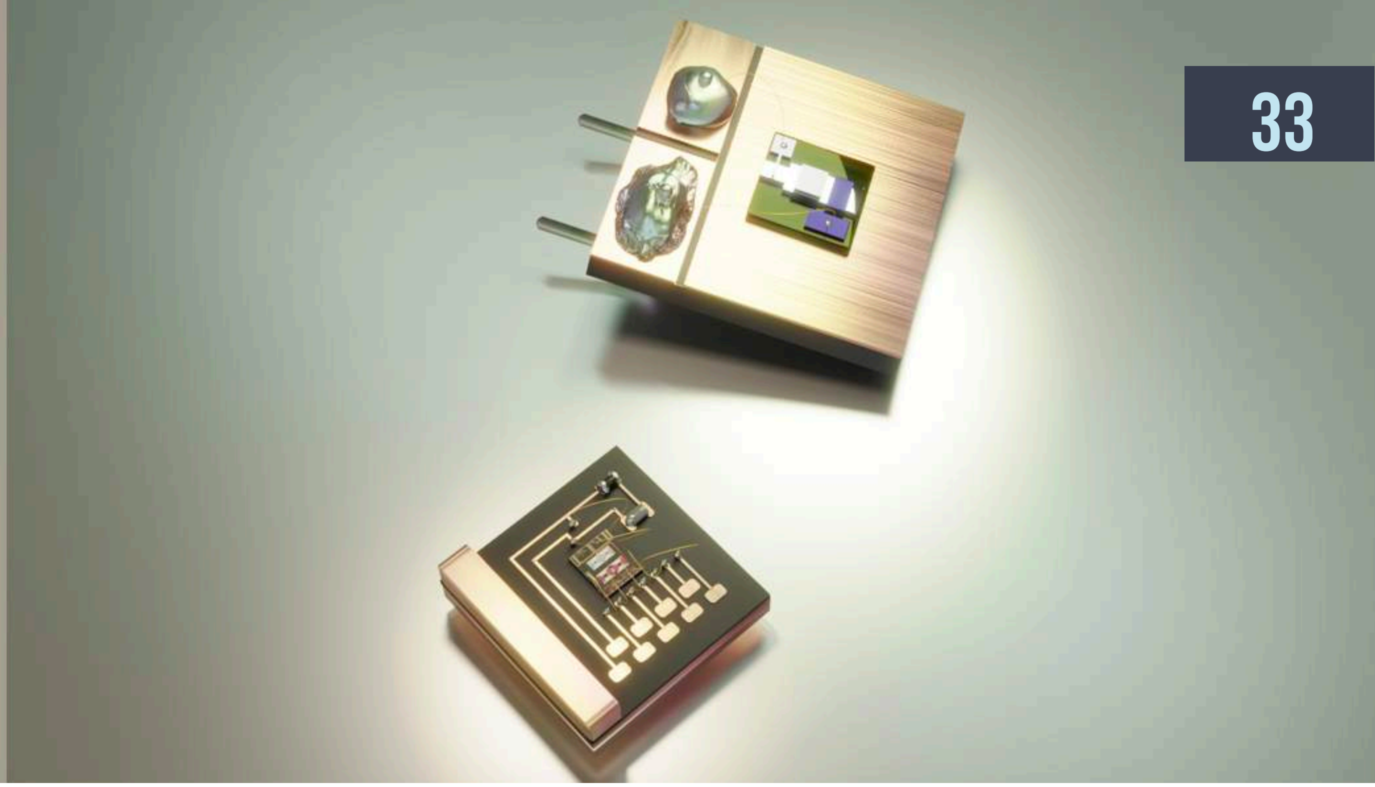


DEEP CRYOGENIC SIGNAL READ-OUT

Superconducting sensors provide the highest performance, but with the price of working in complex environments which make the handling of the signals and overall operation rather complex. The main reason for this is the need for deep cryogenic temperatures required for reaching the superconducting state of some of the materials used in the multilayered structure. The extraction of the signal from cryostats requires a careful design of the read-out system. Furthermore, high-end applications typically imply detection of very low signal levels, and therefore the aforementioned read-out will require some type of signal amplification.

All these aspects were very carefully studied during the SUPERTED project. Theoretical models have shown that the sensors based on FI/S structures can reach at least as good, or even better, figures of merit than Transition Edge Sensors (TES), which is one of the superconducting sensing technologies that currently sets the level of high performance. These models also allowed us to understand that (low temperature) voltage amplification stages are better adapted than current amplification stages, because the amplification noise of the former is below that of the noise of the sensor itself, while the latter will induce levels that overcome the





First level of integration: The sensor is fixed to a platform with two electrical contacts, which allow extracting the output signal. The connection between the sensor and the larger electrical contacts is done by means of Au wires. The electrical contacts link with the first stage of the read-out, consisting of an LC circuit, coupled to the SQUID.

signal at low temperatures. Such conclusion is, however, not easy to implement in practice because of the lack of suitable voltage amplification technology, given the operational constraints.

If the sensor has been correctly optimised, and therefore the transducer matches the impedance of the antenna, then the size of the junction is small, and dynamic impedance may rise to the level of several kΩ or even MΩ. This leads to very high levels of noise for voltage amplification based on, for instance, bipolar transistors. Other techniques, better suited for high-impedance devices, like Junction Effect Transistors (JFET) or AlGaAs/GaAs heterojunction Field Effect Transistors will unfortunately not be easy to adapt because of the required operating temperature and the power dissipated, which are difficult to handle inside the cryogenic environments.

The theoretical study of an electromagnetic radiation detector based on the Giant Thermoelectric Effect can be found in the following publication:

•*"Thermoelectric Radiation Detector Based on Superconductor-.ferromagnet Systems"* Phys. Rev. App.. **10**, 034053 (2018). DOI: doi.org/10.1103/PhysRevApplied.10.034053

The design of the read-out systems is addressed in detail in Deliverable 3.1. "Read-out Scheme".

The discussion in this deliverable is complemented in the following publications:

- *Superconductor-ferromagnet tunnel junction thermoelectric bolometer and calorimeter with a SQUID readout* , J. Low-Temp. Phys. **199**, 585-592 (2020). DOIC: https://doi.org/10.1007/s10909-020-02419-0
- Analytical Models for the Pulse Shape of a Superconductor-Ferromagnet Tunnel Junction Thermoelectric Microcalorimeter, J. Low-T. Phys. **209**, 419–426 (2022), DOI: https://link.springer.com/article/10.1007/s10909-022-02768-y

An alternative read-out based on current amplification using Superconducting Quantum Interference Devices (SQUID) turned to be an attractive option, because of several reasons: 1) it is compatible with deep cryogenic environments, 2) the system dissipates very low levels of heat, 3) typical bandwidths of 10-20 MHz are high enough not to limit the response time of the sensor, and 4) high impedance of the sensor does not induce significant levels of noise in the read-out.

Integration of the sensor and the first stage of read-out: Both devices must be located close to each other, in a zone with the lowest temperature. SQUID read-out must be shielded from the magnetic field applied to the sensor in order to initiate the spin splitting.

The circuit is directly connected to a superconducting flux transformer at the front of the input coil of the SQUID. This helps to reduce the current noise which is otherwise too high for the sensor, and helps achieving a better matching between the sensor and the read-out system.

Integration of both the sensor and the first stage of the SQUID read-out should be done in the deep cryogenic environment (~0.1K), but requires some adaptation of the space.

In current SUPERTED sensors, application of a magnetic field is required to align the magnetic domains of the EuS layer of the transducer and generate the exchange field which is responsible for the spin splitting in the Density of States. This can be done using a Helmholtz coil (i.e., two toroidal electromagnets aligned on the same axis, carrying an equal electric current in the same direction), that creates an almost uniform magnetic field in the volume inside the coil (where the sensor is positioned).

Since the first stage of the SQUID read-out has to be located at proximity of the sensor, it is necessary to shield this circuit from the effect of the applied magnetic field. This is done by placing the SQUID circuit inside a box made of Mu_metal (nickel–iron soft ferromagnetic alloy).

The second stage of the read-out is located in a zone with temperatures of approximately 1.5 K, and consist of a SQUID array coupled to the first stage through an input coil. Both 0.1 mK and 1.5 K stages are surrounded by a 4K zone. The remaining read-out system is located outside the cryogenic environment, and consists of a Pre-Amplification stage coupled to an integrator, and a Digital-Analog Converter.

The aforementioned scheme is in principle applicable to both bolometer type and microcalorimeter sensors, but the behaviour is very different:

- In bolometers the input flux transformer has an impact both in (current) noise and speed of the device. While the thermal time constant of the sensor is independent of the read-out, the electrical time constant is determined mainly by the inductance of the front-end signal coil.

- In micro-calorimeters the absorber, made of heavy elements for efficient absorption of the high energy photons, leads to a much longer thermal time constant of the device, which is normally longer than the electrical time constant of the SQUID. Engineering the transparency of the tunnel barrier helps increasing the sensor bandwidth.

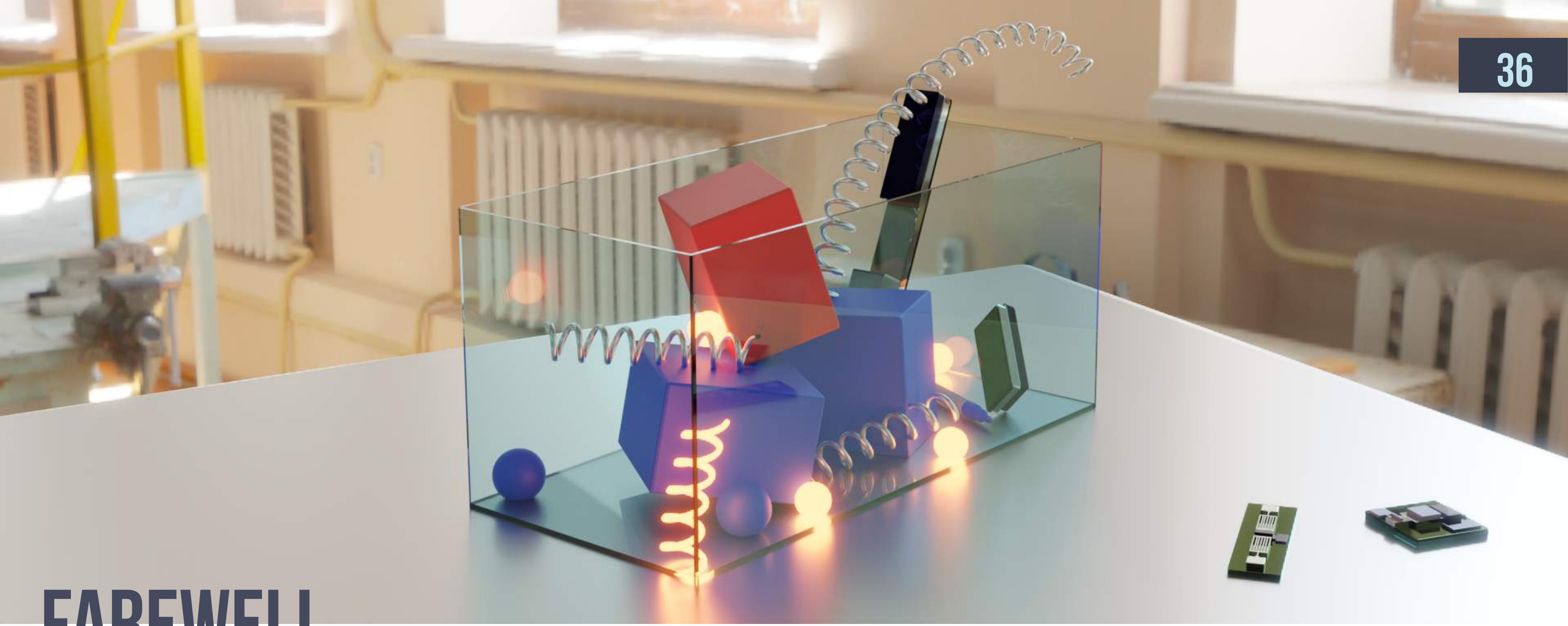
The SQUID read-out can be adapted to be implemented in arrays of sensors, using multiplexing schemes:

- In Time-division multiplexing (TDM) scheme each sensor has a dedicated flux transformer and first stage SQUID read-out. Each row of SQUIDs is biased in series with a timed sequence, and the output signals are collected by the column-wise summing coils and measured by the secondary SQUIDs.

- In Code-division multiplexing (CDM), each pixel is orthogonally modulated by Walsh codes (highly auto-correlated but zero cross-correlated) functions, measured sequentially in each row, and separated at last by a matrix operation. This can be implemented by using input coils for each sensor, connected in series but with different polarisations, and coupled flux transformers and first stage SQUIDs. These SQUIDs are switched on in a timed sequence, and the output signals are collected by a summing coil and measured by the secondary SQUID.

Alternatively, sensors can be adapted to implement KID-like read-outs, as shown in Chapter 3.

The last adaptation of the cryostat systems concerns the opening through which the sensor can be exposed to the incoming electromagnetic radiation. The design of such an opening is highly dependent on the application, as it is composed of a series of windows that isolate the different stages of the cryostat (i.e. zones with different temperatures) and a series of filters designed to eliminate parasitic radiation.



FAREWELL

This is as far as we have gotten. The SUPERTED project has given us the immense privilege and opportunity to take a new discovery and explore its potential.

It is an interesting exercise to compare the progress made in SUPERTED with that of another well established superconducting sensor technology. Transition Edge Sensors (TES) were first demonstrated in 1940, almost 30 years after the discovery of the physical phenomena on which they are based (the sudden drop of resistance when reaching a critical temperature, discovered by Heike Kamerlingh in 1911). The solution of stability problems (thermal runaway problem) that hindered the practical implementation of TES was not found until 1995.

The sensors based on Giant Thermoelectric Effect are, of course, not yet competing at the same level as the TES, but the SUPERTED project has allowed to drastically shorten the times required for realising the first devices and testing them with relevant applications in mind. The usefulness of the European funding is crystal clear in this case. Without it, the different groups of researchers, from very different communities, who contributed to the project would have hardly had the chance to get together and discuss. Instead, they have constituted a world-class team which has made very significant progress and solved difficult technical challenges. It is worth stressing that the Giant Thermoelectric Effect is scientific knowledge, so it may well find other applications and generate more interesting technologies, as some of the publications of the SUPERTED project have already started to show.

We will now keep looking for new opportunities to further develop this technology, and hope to get other researchers involved along the way.

SUPERTED

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