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# STATEMENT

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Ústav fyziky

Teze disertační práce

# Electron beam characterization of technical surfaces at cryogenic temperatures

# Charakterizace technických povrchů elektronovým svazkem za kryogenních teplot

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# Abstract (EN)

This dissertation presents applied research on the electron irradiation-induced emission of electrons and molecules and thermally controlled gas adsorption and desorption at cryogenic temperatures. Various technical-grade metal surfaces and functional surface coatings and treatments are studied under conditions relevant to many technical applications. A particular focus is on understanding the electron cloud and dynamic vacuum phenomena in CERN's Large Hadron Collider (LHC), which operates at cryogenic temperatures below 20 K. Its electron cloud is characterised by low energies in 0–1 keV range but high doses up to 10 mC.mm$^{-2}$. Such conditions are controllably reproduced in a newly developed cryogenic laboratory setup designed for collector-based measurements of Secondary electron emission (SEY), electron stimulated desorption (ESD), and temperature programmed desorption (TPD) at high sensitivity, precision, and accuracy. The experimental results are acquired, analysed and systematically discussed in detail. Finally, semiempirical parametric models of the SEY and ESD yields are developed to capture the energy, dose, angle, temperature and composition dependencies, allowing further use in the field. While emphasising the LHC's electron cloud-induced dynamic vacuum effect and related phenomena, the research findings are interpreted in a generalist manner, making them relevant to other accelerators and technical applications.

**Keywords:** secondary electron emission, electron stimulated desorption, temperature programmed desorption, cryogenic temperatures, cryosorbed gases, technical-grade metals, coatings and treatments

# Abstrakt (CZ)

Tato disertační práce se zabývá aplikovaným výzkumem emise elektronů a molekul vyvolané elektronovým zářením a tepelně řízenou adsorpci a desorpci plynů za kryogenních teplot. Studovány jsou různé technické kovové povrchy a funkční povlaky a povrchové úpravy za podmínek relevantních pro mnoho technických aplikací. Zvláštní pozornost se věnuje jevů elektronového oblaku a dynamického vakua ve Velkém hadronovém urychlovači (LHC), který pracuje za kryogenních teplot pod 20 K, a jehož elektronový oblak má nízké energie v rozmezí 0–1 keV ale vysoké dávky až po $10\,\mathrm{mC.mm^{-2}}$. Takové podmínky lze řízeně reprodukovat v nově vyvinutém kryogenním laboratorním systému určeném pro vysoce citlivé a přesné kolektorové měření sekundární elektronové emise (SEY), elektronově stimulované desorpce (ESD) a teplotně programované desorpce (TPD). Získané experimentální výsledky jsou podrobně analyzovány a systematicky diskutovány. Nakonec jsou vyvinuty semiempirické parametrické modely pro jevy SEY a ESD, které zachycují závislosti na energii, dávce, úhlu, teplotě a složení a umožňují další využití výsledků v této oblasti. Přestože je kladen důraz na efekt dynamického vakua vyvolaný elektronovým mrakem na urychlovači LHC a související jevy, jsou výsledky výzkumu interpretovány obecně, takže jsou relevantní i pro jiné urychlovače a technické aplikace.

**Klíčová slova:**
sekundární elektronová emise, elektronově stimulovaná desorpce, teplotně programovaná desorpce kryogenní teploty, kryosorbované plyny, technické kovové povrchy, povrchové úpravy

# Contents

# Introduction

This dissertation orients on vacuum science and the physics of surfaces under electron irradiation. The manuscript consists of four major parts that aim first to motivate the reader, then state the research niche, guide through the applied research work, and ultimately reconnect to the experimental results' applicability within the applied research field.

The first two chapters introduce the technical context and its state-of-the-art. Careful analysis of the problematics arising from electron irradiation of technical-grade metal surfaces allows for setting experimental goals and methods.The phenomena of secondary electron emission (SEY), electron stimulated desorption (ESD) and temperature programmed desorption (TPD) are investigated with a particular (but not exclusive) focus on understanding the electron cloud-induced dynamic vacuum effect in CERN's Large Hadron Collider, the LHC.

The Experimental chapter describes the setup commissioning, calibration and the methodology developed to deliver quantitative results on SEY, ESD and TPD in the targeted temperature, pressure and energy ranges.

The Results chapter systematically presents the acquired results and immediately discusses the experimental and physics standpoint, transforming data into knowledge. The SEY is addressed first as the origin of the electron cloud activity and is followed by the ESD that links it to the dynamic vacuum effect, both observed in the LHC. The irradiation and environmental parameters are consecutively addressed, allowing to disentangle the influence of the primary electron energy, dose, angle, surface temperature, treatments and cryosorbed gases. The TPD measurements then follow as a means to characterise specific surfaces of treatments and the impact of cryosorbed gases on the SEY and ESD. This chapter intertwines the engineering perspective with the theoretical background necessary for a correct data interpretation.

Finally, the Applications chapter elaborates on the research results' direct applicability. Although the LHC relevance is imperative, the experiments are interpreted in a generalist manner to suit other technical applications. The state-of-the-art theory-backed understanding can be leveraged to generalise the emergent dependencies for further use.

# Chapter 1
# Motivation

CERN's accelerator complex consists of a chain of accelerator rings that circulate and accelerate high-energy proton and ion beams and provide them to various experiments, mainly the LHC. The LHC [1] largely consists of superconducting cryomagnets, whose strong magnetic field focuses and circulates the beam on a closed circular trajectory. Those magnets' cold bore houses a beam tube with a specially designed beam-screen (BS) liner. Supercritical LHe cools the BS to 5–20 K range and extracts heat generated by the circulating proton beam before reaching the superconducting magnets' cold mass, see top-centre of Fig. 1.2

The circulating high-intensity bunched proton beam emits synchrotron radiation [2] that extracts photoelectrons from the beam-screen that further multipact into an electron cloud (EC) as schematised in Fig. 1.1. The EC activity is determined mainly by the secondary electron yield (SEY), which is an energy-, dose- and angle-dependent characteristic giving the average number of emitted electrons per one impinging electron. These electrons then degrade the ultra-high beam vacuum by releasing gas from the beam-screen surface via the electron stimulated desorption (ESD) phenomenon. Both SEY and ESD are extremely surface-sensitive parameters.

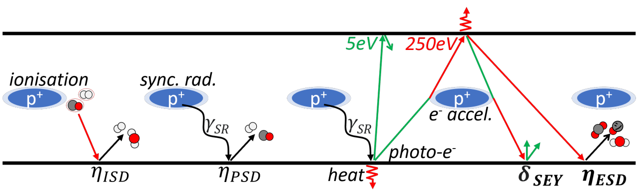


Figure 1.1: Main interactions between the proton beam, residual gas and the beam tube surface: the bunch-per-bunch electron-cloud build-up and the ion-, photon-, and electron-stimulated gas desorption mechanisms.

The beam-screen is designed [3] to mitigate the electron cloud and maintain the ultra-high vacuum even when circulating proton beams. Fortunately, both the SEY-driven EC activity and the ESD-induced dynamic vacuum effect decrease with time in operation as the BS conditions under high irradiation doses. However, the electron cloud activity and the heat dissipated into cryogenics are still limiting factors for the efficient operation of the LHC and its upcoming upgrade, the High-luminosity LHC.


Figure 1.2: Outreach of this applied research work to other applications and domains. Ranging from engineering applications and surface analysis, past high-energy physics, to astrophysics: all dealing with electron irradiation of, possibly cold, ill-defined surfaces.

Electron cloud and dynamic vacuum effects were observed in many other storage rings circulating bunched charged particles, such as: RHIC in the USA [4] with its electron-ion variant [5], present and future GSI machines such as SIS100 [6], and SuperKEKB in Japan [7, 8], including light-sources [9]. Pressure rise by up to ∼5 orders of magnitude are documented, which is far beyond acceptable. Available problematics overviews, e.g. [10], including simulations [11] and measurements [12] show that EC energy distribution mostly resides in the low energy-range, with a major peak below 10 eV. When the electron multipacting occurs, the major peak is followed by a secondary peak at a few hundred eV. Many solutions were proposed to mitigate these effects, including the carbon-coatings [13, 14] and laser-treatments [15–17] investigated here.

In parallel to haunting accelerators in operation, the ESD-induced pressure rise in vacuum devices is a long-standing technical challenge. Ranging from vacuum gauges and mass spectrometery, where it produced a dynamic background signal, to vacuum tube technology [18, 19]. Hence, electron irradiation, ill-defined technical-grade surfaces, often made of metal, and subsequent emission of secondary electrons and gas desorption are common under many scenarios and different conditions. The application fields range from astrophysics [20, 21], or radiation damage to biological samples [22], past EC formation in radiofrequency devices [23–25], towards impurity influx control in plasma devices [26–28].

# Chapter 2
# State of the Art

This literature survey covers the relevant phenomena: secondary electron yield (SEY), electron stimulated desorption (ESD) and temperature programmed desorption (TPD). Each section contains a brief theoretical reminder and a survey of relevant experimental data. The review starts out with the various electron-matter interactions that are a prerequisite for secondary electron emission and gas electrodesorption, see Fig. 2.1.

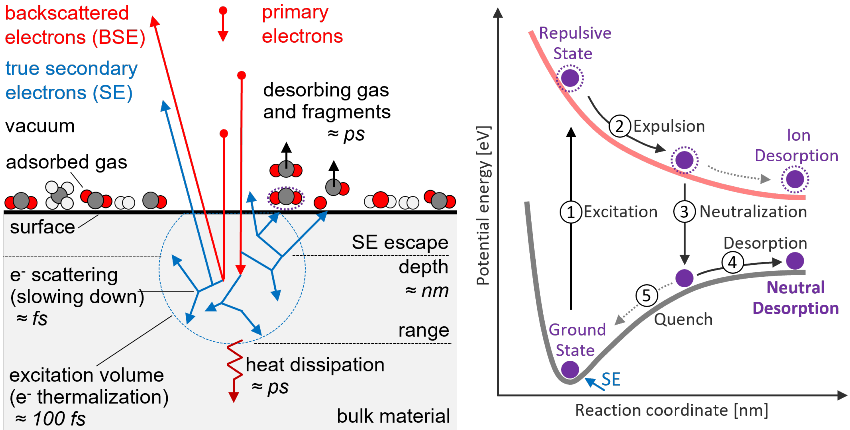


Figure 2.1: Left: Electrons interacting with a technical-grade surface, leading to the emission of secondaries. Right: Schematised and simplified DIET sequence leading to neutral desorption. The main and alternative routes are visualised. The scheme is illustrative and not to scale. Credit: Author.

This critical up-to-date overview of state-of-the-art covers the physical phenomena underlying sub-keV electrons' interaction with technical-grade metal surfaces held at cryogenic temperatures. The effect of various parameters on the ESD, SEY and TPD is discussed and numerous examples illustrate the main trends and numerous unknowns, forming research niches. This includes material and surface properties, irradiation characteristics and environmental conditions.

### 2.0.1 Secondary electron yield

The Secondary Electron Yield (SEY), denotes $\delta$ and defines as the average number of electrons leaving the surface per impacting primary electron. Since the SEY is the main parameter driving the electron-cloud activity in the LHC cryodipoles, it is closely followed in the manuscript. This research covers the SEY in 0-1.5 keV energy range that has a number of distinct features. Particular focus is put on 0∼40 eV range, as it is representative of the electron cloud energy spectrum irradiating the BS.

$$\delta\ [e^-/e^-] = I_{Secondary}/I_{Primary} \tag{2.1}$$

### 2.0.2 Gas desorption induced by electron impact

A range of mechanisms can lead to a gas species emission due to electron irradiation, see Fig. 2.1. Each has a distinct signature behaviour and many are observable in this research. The quantity followed here is the electron stimulated desorption (ESD) yield, defined as an average number of gas molecules desorbed by an impinging electron:

$$\eta_j\ [molecule/e^-] = \frac{C_j.\Delta p_j}{k_B.T} \Bigg/ \frac{\Delta I_b}{q_e} \tag{2.2}$$

This literature survey amalgamates the available data and theory to form an understanding and bridge numerous blank spaces. While many educated guesses can be made for a particular system regarding its ESD yields, thresholds, and conditioning rates, true predictability in an extrapolative manner is still missing. This is addressed in the coming chapters.

### 2.0.3 Temperature programmed desorption

To measure the ESD and SEY of technical surfaces covered with cryosorbed gases, one needs a way to characterise the substrate-adsorbate system. Temperature programmed desorption (TPD) is an experimentally accessible method that essentially measures gas desorption during a linear temperature ramp-up. The TPD curve shape depends on the underlying physical processes and can be described by the Arrhenius rate equation:

$$r\ [mbar.l^{-1}.s^{-1}.cm^{-2}] = -d\theta/dt = \nu\ \theta^n\ \exp(\frac{-E_{ads}}{k\ T}) \tag{2.3}$$

The shape reveals much about the substrate-adsorbate interaction in terms of the adsorbate coverage, regime, binding energy, adsorption site distribution, specific surface, etc. [29–31].

# Chapter 3
# Research objectives

## 3.1 Research niche identification

The Motivational chapter underlines the relevance of the studied problematics and the need for research results. The survey done over commonly accessible literature demonstrates that standard DIET models developed over the past decades provide a coherent view of the ESD problematics. The DIET treatment delivers good qualitative predictions in agreement with experimental observations presented in available ESD literature. However, obtaining precise quantitative predictions using the theoretical approach is challenging, even for well-defined systems. This task is margining the impossible when predicting the ESD properties of technical surfaces.

The literature survey also shows a great number and variety of data spread over various research fields, including vacuum technology, high-energy physics, astrophysics, radiology, etc. However, the accessible data is still very scarce compared to the vastness of all the parameters influencing both the SEY and ESD. Aside of data scarcity, the situation is made worse by a lack of reliability and overall coherence. It is common to see authors struggling to reproduce the work of others and obtaining results that vary greatly from one author, laboratory, or experiment to another. This is partly because of the lack of an exhaustive description of relevant experimental conditions in terms of process parameters, surface characteristics, purity of compounds, etc. and partly the lack of coherent and standardised terminology used to describe ESD when approached from different disciplines. These deficiencies mark numerous niche areas within the ESD problematics that are to be developed and filled with experimental evidence and knowledge derived from within. In summary, the research field's status is unsatisfactory in terms of available datasets for common technical-grade materials, especially at low energies below 100 eV, and at cryogenic temperatures around 10 K.

## 3.2 Dissertation outline and objectives

### 3.2.1 Dissertation objectives

The dissertation aims to advance knowledge about secondary electron emission and electron-stimulated desorption phenomena at cryogenic temperatures from technically important materials used, for example, in vacuum apparatuses of particle accelerators, electron microscopes, and radiofrequency or plasma devices. The detailed literature survey revealed the absence of knowledge about the molecular emission from technical materials (e.g. Cu, Al and stainless steel) for low electron energies and cryogenic conditions. In addition, there are no suitable experimental data and predictive models that approximate the dependence of molecular emission (as measured by the ESD yield) as a function of environmental, irradiation and surface parameters. Consequently, this research focuses on the study of electron-stimulated desorption for selected low energies and doses of electrons, substrate and adsorbate compositions and temperatures, as well as its relation to the electronic emission yield (SEY parameter). Hence, the research objectives define as follows:

- Definition, development, commissioning and optimisation of experimental methodology relevant to studying SEY and ESD phenomena and characterising technical surfaces at low electron energies and cryogenic conditions.
- Investigation of electronic and molecular emission (as measured by SEY and ESD yields) from selected materials and treatments in the low-energy range 0–1 keV as a function of electron energy, electron dose and substrate temperature.
- Development of parametric models that approximate the acquired experimental dataset as a function of electron energy and dose, substrate temperature, and adsorbate composition.
- Analysis of applicability of the new experimental evidence, developed models and derived knowledge with regard to representative technical scenarios.

These research objectives are discussed in detail in the below paragraphs and the achievement of these objectives is systematically presented in Chapters 4, 5 and 6, respectively.

#### Development of the experimental methodology

In the absence of experimental data, it was decided to device an experiment to study the electron irradiation of common technical materials, especially at low energies below 100 eV, and at cryogenic temperatures around 10 K. It was therefore decided to develop an experimental system and corresponding methodology to perform systematic studies of the SEY and ESD phenomena with precise control of measurement conditions. In particular, those representatives of the LHC and other relevant fields of technology, engineering and science.

### Acquisition of a systematic dataset

We can define the following circles of parameters that align with the thesis title *Electron beam characterisation of technical surfaces at cryogenic temperatures*. That is the influence of material, irradiation and environmental properties onto electron irradiation- related parameters SEY and ESD. The material properties encompass the bulk material, the surface state and possibly the microgeometry. The parameters of electron irradiation are described by their energy, flux, total dose and incidence angle. Finally, the environmental factors should be well described by the substrate temperature, which governs the thermal motion, and by the residual gas composition and by the cryosorbed gas species. Hence, the parameter space of factors influencing the SEY and ESD can be categorised as follows:

- Material properties
  - Bulk material
  - Surface condition
- Irradiation properties
  - Primary electron energy
  - Absorbed electron dose
  - Incidence angle
- Environmental properties
  - Temperature
  - Cryosorbed gases

### Experimental results interpretation, generalisation and application

The research goal is not only to provide a dataset, but also to disentangle the complex interplay of various factors listed in the previous section. The objective is to expand, in a structured manner, the current understanding of ESD problematics into a more coherent and holistic view.

The acquired data on ESD can be processed in the following ways. First, the SEY and ESD measurements are to be critically analysed and compared to theoretical expectations, simulations results and other authors who performed similar measurements. Second, the conditioning and energy-dependence curves of ESD yields can be fitted with suitable models that will provide some degree of predictive capability. This step is particularly important, as models are crucial for further application in numerical simulations of dynamic vacuum. Electron-induced chemistry and transient dynamics can be investigated for metals, treatments, coatings and cryosorbates. Tracing with isotopically labelled molecules could be employed for pure cryosorbed gases and binary mixtures. These can then be compared to the current understanding, gas-phase ionisation data, and other experimental data. Lastly, the precision and accuracy of our experimental approach can also be evaluated for comparison to other measurement methods used in other laboratories.

## 3.3 Impact of this research

The newly developed and commissioned experimental setup and methodology enables an on-demand electron beam characterisation of technical surfaces, coatings and treatments under the irradiation and environmental conditions representative of the LHC cryomagnets. The resulting understanding of SEY and ESD phenomena at cryogenic temperatures should benefit the accelerator design, operation and general research community. The improved understanding of technical materials' vacuum performance at cryogenic conditions will allow for a well-informed choice of construction materials. Surface qualification before an in-situ deployment, post-mortem or witness-sample analyses are also an advantage. Aside from the above-listed technical applications, studying electron irradiation of condensed mater (albeit much thicker) is also relevant to astrophysics community.

# Chapter 4
# Experimental methods

## 4.1 Experimental setup description

Fig.4.1 captures the experimental setup designed to study the ESD and SEY at cryogenic temperatures. It consists of a $\mu$-metal vacuum chamber, a 4-axis cryomanipulator, a low-energy electron gun and a Residual Gas Analyser (RGA) fitted inside a collector. The setup is further equipped with a storage and a load-lock chamber to introduce unbaked samples into the baked experimental UHV chamber, allowing to study samples in an as-received surface state. The chamber and cryomanipulator are custom-built by external manufacturers, whilst the collector and the gas dosing tube are built in-house. The setup layout, setting and experimental procedures were iteratively optimised to deliver the presented data.

The collector' is fundamental to the chosen measurement layout, as it forms a closed geometry that captures secondary electrons and desorbed gases, essentially combining Faraday and Feulner cap functionality.

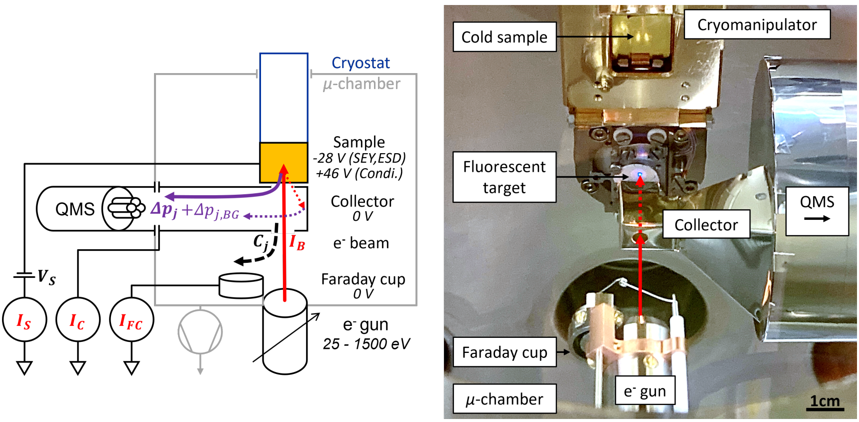


Figure 4.1: Arrangement of the experimental setup specifically designed for low-energy and low-temperature SEY, ESD and TPD measurements. Presented in [A3, A7, A8].

## 4.2 SEY and ESD yield measurement

A procedure was developed for the energy-dependence measurement to collect discrete datapoints at increasing energy, as shown in Fig. 4.2 by a proof-of-concept measurement. The procedure are similar for the SEY and ESD except for different beam and timing parameters. While the SEY is nondestructive at its 0.5 nA beam current, an ESD measurement is in principle destructive at its $\sim 2\,\mu A$ beam current.

The top-left plot in Fig. 4.2 shows the beam current modulation and the sample and collector current varying in response to the changing energy. The bottom-left plot depicts the RGA currents of monitored masses 2, 28 and 30 m/q that vary in response to the square wave-modulated electron beam. The dynamic background, originating at the collector, not the sample, is visualised and marked in the figure and imposes a detection limit, particularly on $H_2$ and CO. The $^{15}N_2$ was selected as it exhibits a high SNR and low background, compared to $H_2$ and CO. This is partly due to a high ESD yield (common to cryosorbed gases) and low background at the 30 m/q channel. The observed threshold are similar to those of Rakhovskaia et al. for $^{15}N_2$ [32] and Billard et al. for $H_2$ and CO (extrapolated) [33], which demonstrates the correct commissioning. The developed experimental methodology is presented in [A3, A7, A8].

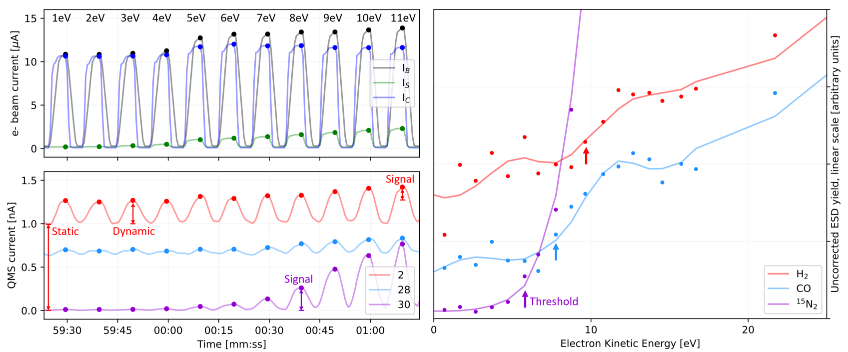


Figure 4.2: Low-energy ESD yield measurement of semi-conditioned Cu held at 15 K with a 1 ML precoverage of $^{15}N_2$ used as a tracer. Top-left: Time series of $e^-$ beam, sample and collector currents, all modulated to a square wave by gating via the grid. The kinetic energy is incremented by 1 eV each cycle. Bottom-left: Filtered RGA currents for channels 2, 28 and 30 m/q modulated in response to the grid-gated $e^-$ beam current. Right: Datapoints and trendlines for $H_2$, CO and $^{15}N_2$ yields as a function of primary $e^-$ kinetic energy. Arrows mark the desorption threshold energies for each gas. Note the noise and dynamic background levels of $^{15}N_2$ compared to $H_2$ and CO. Presented in [A3, A7, A8].

The reference SEY curve measured on a highly-oriented pyrolytic graphite

(HOPG) sample exhibits a fine structure, especially in the low-energy region, see left side of Fig.4.3. Since this curve's signature features are similar to other established surface laboratories, this measurement demonstrates the energy resolution and absolute accuracy across the studied energy range, proving the commissioning correct.

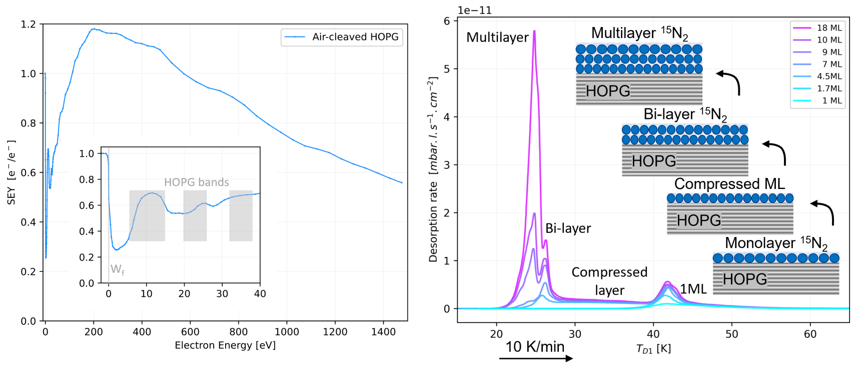


Figure 4.3: Left: SEY curve measured with Multisystem in the 0-1.4 keV range on a HOPG sample held at room temperature. This HOPG sample was exfoliated in air with an adhesive tape and then electron conditioned for contaminant removal. Data presented in [A3, A7, A8]. Right: TPD curves of $^{15}N_2$ quench-condensed on an A-grade HOPG reference sample at 11 K, as exfoliated under vacuum [34].

## 4.3 TPD measurement

Given that cryogenic temperatures bring about inevitable residual gas adsorption on the studied surface, a method of characterising the adsorbed gas adlayer is necessary. Temperature Programmed Desorption (TPD) is an experimentally accessible quantitative method that utilises the combination of an absolute-calibrated Residual Gas Analyser (RGA) and the sample temperature reading.

The method was proven by taking a series of TPD curves of $^{15}N_2$ cryosorbed in 0-18 ML precoverages over the HOPG sample, see right side of Fig. 4.3. The absolute resolution over 5 orders of magnitude, the level of detail on the temperature scale and the general repeatability, coherence, and similarity to other authors, demonstrate the correct commissioning and calibration of temperature and gas load measurement.

To summarise, all the necessary measurement methods were successfully commissioned and deliver SEY, ESD and TPD in agreement with other researchers and laboratories. As such, the setup can be used for further research.

# Chapter 5
# Results and Discussion

## 5.1 SEY of technical surfaces

A wide range of samples was studied to disentangle the influence of electron irradiation properties and surface characteristics on the SEY in the 0–1.5 keV energy range. A variety of surface and bulk compositions, treatments, coatings and surface states was covered between the ambient and cryogenic temperatures. Conditioning with different primary electron energies was also done to demonstrate the effect of electron energy on the SEY conditioning efficiency (and ESD, as shown below, 5.2).

Figure 5.1 shows a set of SEY curves across the 0–1.5 keV energy domain of interest, as measured for various surfaces, or surface states. The figure also includes a zoom (left) into the low-energy region, 0–40 eV which is very important in technical applications and only recently gained recognition in the available literature in the past decade for its direct application to the electron cloud build-up [35].

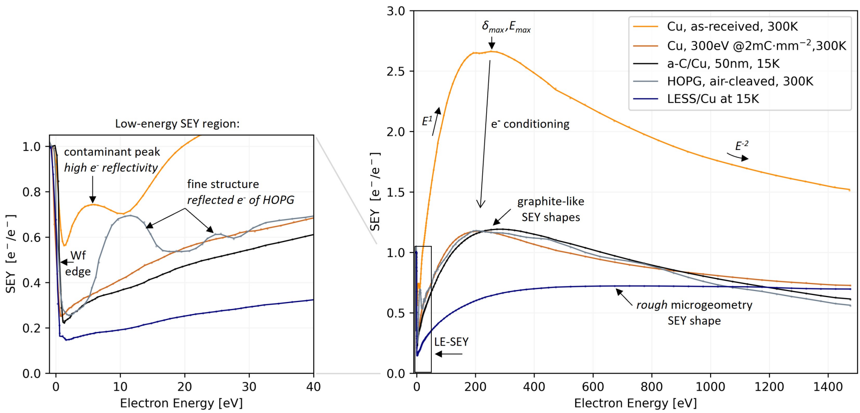


Figure 5.1: Comparison of SEY curves of various surfaces (Cu, Carbon coating, HOPG), surface states (as-received and conditioned Cu) and laser treatment (rough microgeometry). Right: Entire SEY curve in the 0–1450 eV energy region. Left: Zoom into the low-energy region, 0–40 eV. Data presented at [A2, A5, A7].

The following aims were typically pursued. First, to characterise the SEY and ESD under conditions representative of a given sample's technical application, bearing in mind the technical aim of this applied research field. Second, to shed light on intricate dependencies observed in some samples, to improve the understanding and generalise the results. Often to prove a point and support a newly formed hypothesis regarding a specific behaviour - an approach especially useful in the section dedicated to SEY and ESD of cryosorbed gases.

## 5.2 ESD of technical surfaces

The ESD yields energy and dose dependencies were measured for technical-grade Cu, representative of the LHC beam-screen surface, and many other surfaces, treatments and coatings, all relevant to technical applications. Fig. 5.4 on the left shows thresholds at low energy around 10 eV, and decays at high energy above 1 keV, which are qualitatively in agreement with the theoretical understanding.

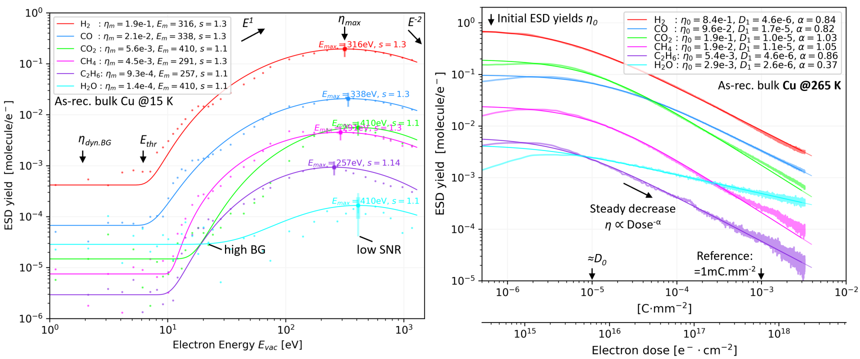


Figure 5.2: ESD yield energy-dependence (left) and dose-dependence (right) measured at 15 K on an LHC-grade Cu colaminated on stainless-steel, in an as-received supplier-cleaned state. Combined uncertainty intervals at $1\sigma$ are $\sim$30 % as shown at the peaks. Data presented in [A2, A6, A7].

Extended exposure to electron irradiation, known as electron conditioning, changes the surface state by removing or reprocessing surface-bound contaminants and inducing diffusion, as shown on the right. These electron-induced changes cause the SEY to decrease, which is a remedy for the LHC's electron cloud issues. Such conclusions were verified using an ex-situ XPS analysis and CASINO electron-tracking simulations, see Fig. 5.3. Note how the deposition depth increases with energy, but the escape depth remains, giving rise to the peaked curve, qualitatively agreeing with the measured data in Fig. 5.4.

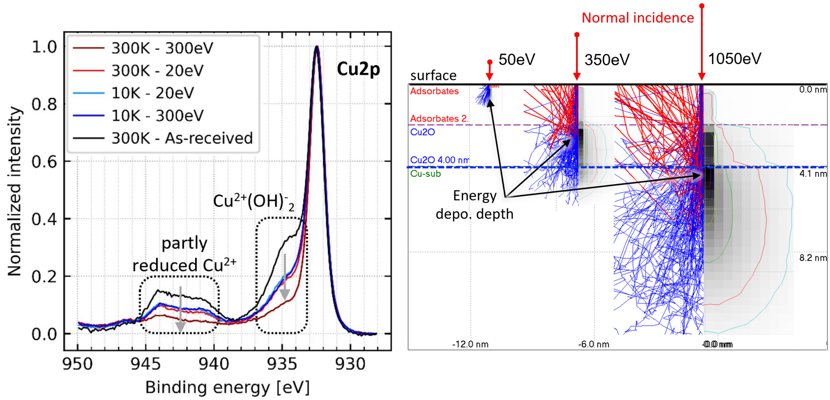


Figure 5.3: Left: XPS (Cu2p line) of LHC-grade Cu in various electron conditioning states shows how the oxidised surface gradually reduces and graphitizes, decreasing the SEY. Right: CASINO simulation [36] of a Cu surface with a contaminant overlayer. Presented in [A3, A5, A7]. Similar analysis is done in [A1].

## 5.3 TPD, SEY and ESD of adsorbates

The cryogenic environment is known for gases cryosorbed on the cold surface from the residual gas. The TPD is a means to characterise the surface and the substrate-adsorbate interaction. Fig. 5.4 on the left shows a TPD of $^{15}N_2$ quench-condensed on Cu/SS substrate in 0.1-11 ML precoverages. Meanwhile, the energy dependence of SEY and ESD yield (right) is important for the electron cloud and dynamic vacuum effect in the LHC's beam-screen, respectively. Note the developed parametric approximation to the ESD data that models the energy threshold, peak and decay.

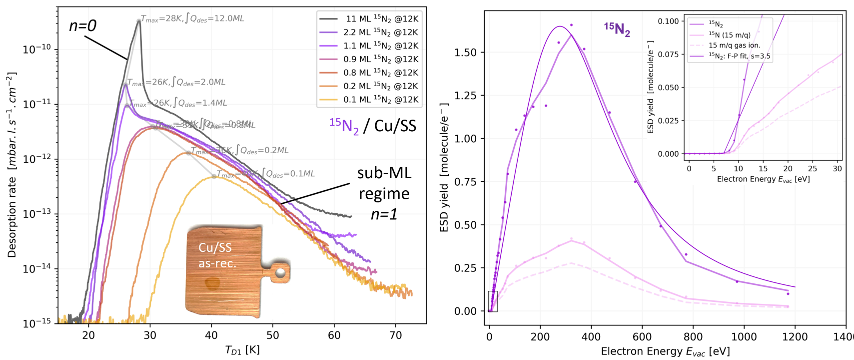


Figure 5.4: Left: TPD of $^{15}N_2$ quench-condensed on Cu/SS substrate in 0.1-11 ML precoverages. Note the change around 1 ML. Right: ESD yield energy dependence for 10 ML of $^{15}N_2$ on Cu, measured as follow-up of [A4]. Note the parametric fit, the 7 eV energy threshold (matching Rakhovskaia [32]) and the fragmentation ratio different from the gas-phase ionisation (as measured or retrieved from NIST [37]).

# Chapter 6
# Applications of research results

## 6.1 SEY and ESD energy and dose models

The parameterised dependencies of the ESD yield on the energy $E$ and dose $D$ are combined into a approximation of $\eta(E, D)$. The ESD yield energy and dose dependencies consist of an initial value, $\eta_{0,max}$ and scaling functions $f(E)$ or $f(D)$ that modulates the input value of $\eta$ with the desired variable $E$ or $D$, as follows in the equation and left side of Fig. 6.1. Only the $H_2$ and CO ESD yields are displayed to illustrate the 2D envelope. Yet, the other common residual gases behave similarly, as seen on the right, see Fig. 5.4. This form correctly approximates the trends observed across many measurements on various surfaces, making it universally applicable, say for calculations and simulation purposes.

$$\eta(E, D) = \eta_{max,0} \cdot \exp\left(\frac{-\ln^2((E - E_{thr})/E_{max})}{2s^2}\right) \cdot \left(\frac{D + D_1}{D_0 + D_1}\right)^{-\alpha} \quad (6.1)$$

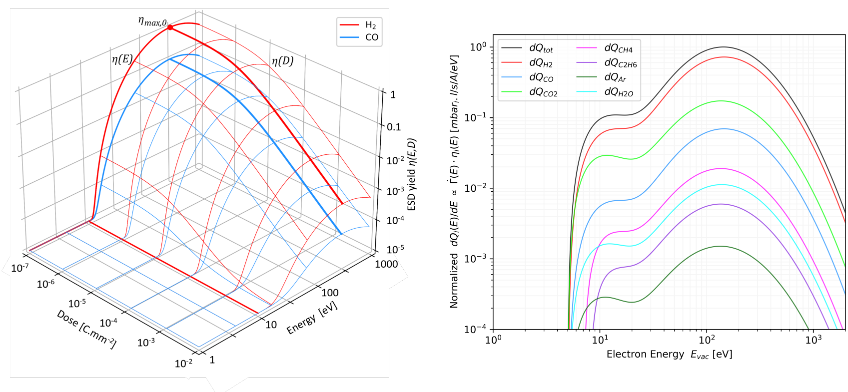


Figure 6.1: Left: ESD yields $\eta(E, D)$ for $H_2$ and CO calculated as a function of energy and dose using the developed approximative model. Right: Energy-resolved gas load for all main residual gases due to electron cloud: 10:1 multipacting peak intensity (True secondary $e^-$ vs. beam-accelerated $e^-$) corresponds to 1:10 gas desorption for beam-accelerated $e^-$. Presented in [A5].

## 6.2 Energy-resolved dynamic gas load

Having the analytical expression for the measured ESD energy dependence $\eta_e(E)$, and an analytical fit to the electron cloud energy spectrum $dN/dE$, the energy-resolved gas load contribution $dQ/dE$ can be calculated for each slice of the energy spectrum $dE$, Fig. 6.1. Note that for a 10:1 ratio of true secondaries vs. beam-accelerated electrons, the gas load already is 1:10 dominated by the beam-accelerated electrons. So the beam-accelerated electrons are some 100x more efficient in desorbing gas than true secondary electrons, which underlines electron cloud mitigation importance.

## 6.3 SEY and ESD of cryosorbed mixtures

A strong departure from a linearly-weighted behaviour is observed for both SEY ($\delta$) and ESD ($\eta$) yields from a 10 ML cryosorbed binary gas mixture, as shown in Fig.6.2. The combining rules were derived for both SEY and ESD to capture this trend by introducing a nonlinear cross-interaction term into equations. The parameters have to be known (and were measured) for pure compounds $A$ and $B$ and can be used to calculate the surface properties of a real cryosorbed gas $A:B$ mixture.

$$\begin{aligned}
\delta_{A:B,max}(x) &= \sqrt[n]{(x-1).\delta^n_{A,max} + x.\delta^n_{B,max}} \\
\eta_{A:B\rightarrow A}(x) &= (x-1)\ .\eta_A + \eta_B\ .x(x-1)\ .\frac{\eta_B - \eta_A}{\eta_A + \eta_B}\ .k^{sign(\eta_B - \eta_A)} \\
\eta_{A:B\rightarrow B}(x) &= x\ .\eta_B + \eta_A\ .x(x-1)\ .\frac{\eta_A - \eta_B}{\eta_A + \eta_B}\ .k^{sign(\eta_A - \eta_A)}
\end{aligned} \tag{6.2}$$

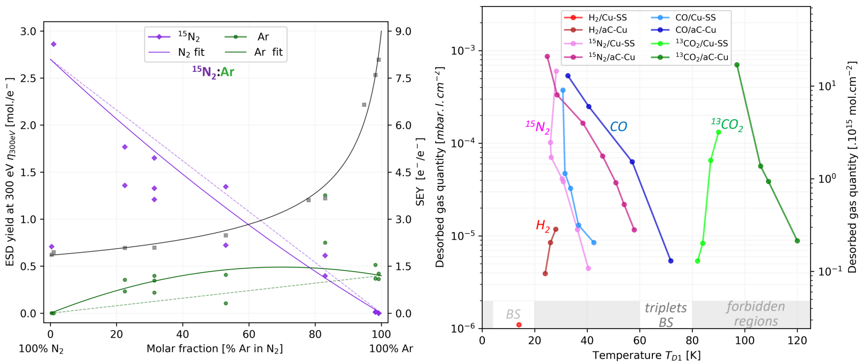


Figure 6.2: Left: $ESD_{300eV}$ and $SEY_{max}$ measured for $N_2$:Ar binary ice at ~10 ML precoverage. The interaction coefficients are $k$=0.5 and $k$=4 for ESD and SEY, resp. Presented in [A5]. Right: TPD integrals for 0.1-10 ML coverages on bare Cu and 50 nm carbon-coated one.

## 6.4 TPD and beam-screen temperatures

Right side of Figure 6.2 shows a TPD integrals measured for typical residual gases adsorbed on LHC BS-representative technical-grade Cu and carbon-coated Cu surface. The surface capacity $Q_{ads}$, peak desorption temperature $T_{max}$ and the binding energy $E_{ads}$ are higher for the carbon-coated Cu, possibly offsetting the suitable operating temperature range, as marked in white/grey at the bottom.

# Summary and conclusions

## Summary of the dissertation

This dissertation presents an applied research in the vacuum science of physical phenomena underlying the to the electron cloud and the dynamic pressure rise observed when circulating the bunched proton beams in the CERN's LHC. This mainly includes the secondary electron yield (SEY), the determining parameter for the electron cloud (EC) activity and possible electron multipacting. The electron stimulated desorption (ESD) yield then determines the amount of gas electrodesorbed due to the electron irradiation coming from the EC. The temperature programmed desorption (TPD) then serves to characterise the amount of cryosorbed gas, its binding energy and the specific surface of the substrate.

To begin with, Chapters 1 and 2 review the underlying problematics, operational observations, simulations and measurements to aim the research efforts better. Fixing the target parameters and research objective and identifying (many) research niches, a new laboratory-based cryogenic experimental setup is conceived and optimised to reproduce the relevant parameters in a controllable manner. Chapter 4 describes in detail the commissioning and calibration procedure of this setup, essentially to correctly count electrons and gas molecules throughout the setup.

In the Results chapter 5, I tackle the combinatorial explosion by systematically exploring this large parameter space and always varying 1 experimental parameter at a time. Various technical-grade metal surfaces, such as Cu, Al and SS, currently used as a baseline for construction, and functional surface coatings and treatments were studied under application-relevant conditions. The SEY and ESD yields of cryosorbed gases and their binary mixtures were also investigated in the 0-30 ML range as they are omnipresent in devices operating under cryogenic conditions. Finally, the Applications chapter 6 presents an overview of semiempirical parametric models developed or modified to capture the SEY and ESD yields dependence on energy, dose, angle, temperature and composition for the studied surfaces, treatments and cryosorbed real gas mixtures. In spite of a particular focus placed on elucidating the electron cloud and dynamic vacuum phenomena in the LHC, the research results are interpreted rather generally to remain relevant to other technical applications. Ultimately, this dissertation provides a state-of-the-art theory-backed yet comprehensive understanding of the physical phenomena underlying the low-energy electron-induced emission of electrons and molecules from technical-grade metal surfaces and functional treatments.

# Conclusions of the research

A particular focus is on understanding the electron cloud and dynamic vacuum phenomena in the LHC, which is characterised by cryogenic temperatures below 20 K [1], low electron energies in 0–1 keV range, especially below 20 eV [10–12], and high electron doses up to 10 mC.mm$^{-2}$. Supporting data were collected in this parameter range of energies, doses and at temperatures that directly envelope the LHC's beam screen operating conditions. Whatsmore, the dataset was often extended to better understand the underlying behaviour and/or broaden the applicability outside the LHC scope.

The electron condition effect was studied and confirmed at cryogenic temperatures for the first time. The presented data, backed by XPS analyses, clearly demonstrate that electron conditioning works equally well at cryogenic temperatures, including electron-induced graphitization of surface-bound carbon-containing contaminants. Moreover, cryosorbed gases tend to fragment intensely under electron irradiation, reprocessing the parent molecules into fragments with vastly different electrochemical and vacuum dynamics behaviours.

Having studied the SEY, being at the electron cloud's origin [38], and the ESD, giving rise to the dynamic vacuum effect [39], the attention shifted to cryosorbed gases influence on SEY and ESD. The SEY of metal surfaces, particularly the low-energy SEY region change already at deeply submonolayer cryosorbed gas coverages, which is particularly relevant to the electron cloud build-up in the LHC [11, 40]. Both increase and decrease in SEY are observed, depending on the particular substrate-adsorbate system. Finally, the SEY and ESD measured on cryosorbed binary gas mixtures exhibited a highly non-linear composition-dependent behaviour that was captured by newly developed combining rules. This is an important step towards realistic extrapolative predictions for real-world scenarios where multi-compound residual gases often co-cryosorb.

The measured SEY and ESD data were approximated by a series of semiempirical models that allow grasping the underlying trends and interpolating across the scarcely populated dataset. Generalising the experimental observations and grasping the emergent patterns is an important step in refining the understanding and generating knowledge from the acquired data. The derived semiempirical parametric models that approximate the SEY and ESD allow grasping the underlying trends, interpolating across the scarcely populated datasets, or coupling the existing electron-cloud simulations [35] to vacuum dynamics calculation software [41].

# Assessment of achieved dissertation objectives

The dissertation objectives outlined in Chapter 3 were successfully achieved on all fronts, effectively expanding the current knowledge in the field of science and engineering.

- Innovative experimental methodology was developed and enabled a detailed combined study of low-energy electron impact-induced electronic and molecular emission (SEY and ESD parameters) from technical surfaces at cryogenic temperatures and with adsorbed gases.
- Uniquely detailed study was performed for various technical-grade surfaces held at cryogenic temperatures and resulted in a detailed investigation of the electronic and molecular emission phenomena as a function of material (metals, coatings, treatments), irradiation (electron energy, dose, angle) and environmental (substrate temperature, adsorbate composition) properties.
- Large amounts of acquired data allowed the formulation of parametric models that well approximate the data (both original and external), and generalisation of the findings and observations. This includes parametric semiempirical models that capture the energy, dose, angle and composition dependence of SEY and ESD yields for various technical surfaces, and can be directly used in further research and engineering practice.
- The newly collected SEY, ESD and TPD data and the derived knowledge was immediately confronted with practical applications and scenarios. For instance, linking the electron cloud activity (its energy and dose dependence) to the dynamic vacuum effect in the LHC, or considering suitable temperature windows for beam-screen operation.

## Personal contribution and scientific results

To begin the research, I elaborated an extended literature review of the studied phenomena and surveyed the diverse applications that may benefit from this research. The resulting manuscript of an up-to-date literature review is yet unpublished. While continuously expanding this knowledge background, I proceeded to commission the laboratory-based experiment.

I was privileged to build, commission, calibrate, and iteratively improve the experiment, originally designed by V. Baglin and B. Henrist, based on their extensive prior experience. This innovative top-class analytical *Multisystem* setup is designed for a combined collector-based measurement of ESD, SEY, TPD and possibly others. This multi-domain experimental capability enables unique research and resulted in a number of articles produced in recent years.

The applied research work yielded an experimental technique-oriented article [A3] and an article with the first results [A2], which I both wrote as the main contributing author. Still, as the main author, I led the collaborative work between three institutes on a future-focused article [A1] dealing with applying high-temperature superconductors in high-energy physics. The research work even overlapped to an astrophysics-relevant publication by R. Dupuy et al. from the Sorbonne University [A4], which I co-authored, and deals with the irradiation of thick cryosorbed ices. Given the newly available experimental data, work is underway on a follow-up article focusing on thin cryosorbed gases deposited over technical surfaces.

I further presented the research work at a number of conferences, where it triggered the interest of the scientific community and received an IUVSTA Elsevier Student Award. This includes conferences such as the $16^{th}$ European Vacuum Conference [A7], the $35^{th}$ European Conference on Surface Science [A6], $18^{th}$ International Conference on Thin Films & $18^{th}$ Joint Vacuum Conference [A8], and the 2022 Electron Cloud workshop [A5].

# List of publications

The authored publications and conference participations related to this dissertation topic are marked as [A1]-[A8] in the manuscript.

## First author publications

[A1] Haubner, M., Krkotić, P., Baglin, V., et al. (2023). Electron Beam Characterization of REBCO-Coated Conductors at Cryogenic Conditions. Applied Sciences. 13(5), 2765. DOI: 10.3390/app13052765

[A2] Haubner, M., Baglin, V., Henrist, B. (2022). Electron conditioning of technical surfaces at cryogenic and room temperature in the 0–1 keV energy range. Vacuum, 207, 111656.
DOI: 10.1016/j.vacuum.2022.111656

[A3] Haubner, M., Baglin, V., Henrist, B. (2022). Collector-based measurement of gas desorption and secondary electron emission induced by 0–1.4 keV electrons from LHC-grade copper at 15 K. Nuclear Instruments and Methods in Physics Research - B, 531, 34-43.
DOI: 10.1016/j.nimb.2022.09.013

# Co-authored publications

[A4] Dupuy, R., Haubner, M., Henrist, B., Fillion, J. H., & Baglin, V. (2020). Electron-stimulated desorption from molecular ices in the 0.15–2 keV regime. Journal of Applied Physics, 128(17), 175304.
DOI: 10.1063/5.0021832

# Conferences and workshops

[A5] Haubner, M., Baglin, V., Henrist, B. (2022). SEY and ESD of ices and technical surfaces at cryogenic temperatures. Electron-cloud workshop and Vacuum for gravity wave detectors 2022, ECLOUD'22 and GWD-Vac'22 workshop. URL: agenda.infn.it/event/28336/contributions/177366

[A6] Haubner, M., Baglin, V., Henrist, B. (2022). Analysis and modification of technical surfaces by low-energy electron irradiation. 35th European Conference on Surface Science. Poster presentation. ECOSS-35.
DOI: 10.13140/RG.2.2.30114.09927

[A7] Haubner, M., Baglin, V., Henrist, B. (2021). Electron conditioning of technical surfaces at cryogenic and room temperature in the 0–1 keV energy range. 16th European Vacuum Conference. EVC-16.
URL: secure.key4events.com/key4register/images/client/1090/files/EVC-16-Programme.pdf

[A8] Haubner, M., Baglin, V., Henrist, B. (2020). Preliminary electron desorption results of selected HL–LHC technical surfaces at cryogenic temperature. 18th International Conference on Thin Films & 18th Joint Vacuum Conference. ICTF-JVC 2020.
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