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# **ecx Documentation**

***Release 0.1.0***

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## GETTING STARTED

Sources: <https://github.com/MilanSkocic/ecx>

### 1.1 ecx



*ecx* is a Fortran library providing formulas for electrochemistry with a C API.

#### 1.1.1 How to install

The library is built with `fpm` which will build the fortran code into a static library. An Makefile is provided which uses `fpm` with additional options for:

- copy the library into the python wrapper folder
- install the C headers
- uninstall the library and headers

On windows, `msys2` needs to be installed.

Build

```
source configuration
make
```

Run tests

```
fpm test
```

Install

```
make install
```

Uninstall

```
make uninstall
```

## 1.1.2 Dependencies

```
gcc>=10
gfortran>=10
fpm>=0.7
```

## 1.1.3 License

GNU General Public License v3 (GPLv3)

# 1.2 pyecx

Python wrapper around the [Fortran ecx library](#).

## 1.2.1 How to install

```
.. code-block:: bash

    pip install pyecx
```

## 1.2.2 Dependencies

## 1.2.3 License

GNU General Public License v3 (GPLv3)

# 1.3 Examples

## 1.3.1 Example in Fortran

```
program example_in_f
    use iso_fortran_env
    use ecx
    implicit none

    real(real64) :: w(3) = [1.0d0, 1.0d0, 100.0d0]
    real(real64) :: r = 100.0d0

    print *, ecx_eis_zr(w,r)

end program
```

### 1.3.2 Example in C

```
#include <stdio.h>
#include <stdlib.h>
#include "ecx_eis_capi.h"

int main(void){

    double w = 1.0;
    double r = 100.00;
    size_t n = 1;
    ecx_cdouble z = ecx_cbuild(0.0,0.0);

    ecx_capi_zr(&w, r, n, &z);

    printf("%f I%f \n", creal(z), cimag(z));

    return EXIT_SUCCESS;
}
```

### 1.3.3 Example in Python

```
import numpy as np
from pyecx import eis
import matplotlib.pyplot as plt

R = 100
C = 1e-6
w = np.logspace(6, -3, 100)

zr = np.asarray(eis.zr(w, R))
zc = np.asarray(eis.zc(w, C))
zrc = zr*zc / (zr+zc)
print("finish")

fig = plt.figure()
ax = fig.add_subplot(111)

ax.set_aspect("equal")
ax.plot(zrc.real, zrc.imag, "g.", label="R/C")

ax.invert_yaxis()

plt.show()
```





## THEORETICAL BACKGROUND

### 2.1 Kinetics

#### 2.1.1 Nernst potential

#### 2.1.2 Buttlar-Volmer

### 2.2 EIS

#### 2.2.1 Introduction

Frequency dispersion measurements (or impedance spectroscopy) have become a common technique for the study of mass and charge transport in electrochemical systems. With the availability of automated high quality frequency response analysis systems immittance (i.e. impedance or admittance) measurements can be obtained in fairly easy way [1].

The advantage of measurements taken in the frequency domain over measurements in the time domain (i.e. pulse or step response measurements) is that the frequency response can be described analytically, using an equivalent circuit as model. Time domain analysis often requires the approximation of complex functions, e.g. infinite summations of exponential functions. The circuit elements represent the various (macroscopic) processes involved in the transport of charge and mass. The dispersion relations for most equivalent circuit elements are very simple Barsoukov and Macdonald [1], Orazem and Tribollet [2].

If the (complex) immittance diagrams show distinct features, which can easily be related to specific subcircuits of the equivalent circuit model, analysis become quite simple. Often this can be accomplished by graphical means, using a compass and a ruler. However, if the time constants of the respective subcircuits are close together, or if elements with a fractional (e.g. Warburg, or a CPE-type element, a more sophisticated analysis procedure is needed. As the variation of one circuit parameter can influence large parts of the frequency dispersion, all parameters must be adjusted simultaneously in order to obtain the optimum fit to the data [3, 4, 5]

#### 2.2.2 Black Box Approach

- Assume a black box with two terminals.
- One applies a voltage and measures the current response (or visa versa) as show in Fig. 2.2.1
- Signal can be dc or periodic with frequency  $f$ , or angular frequency  $\omega = 2\pi f$  as shown in Fig. 2.2.2

with:  $0 \leq \omega < \infty$ :

- Voltage:  $V(\omega) = V_0 \cdot e^{j\omega t}$
- Current:  $I(\omega) = I_0 \cdot e^{j(\omega t - \phi)}$

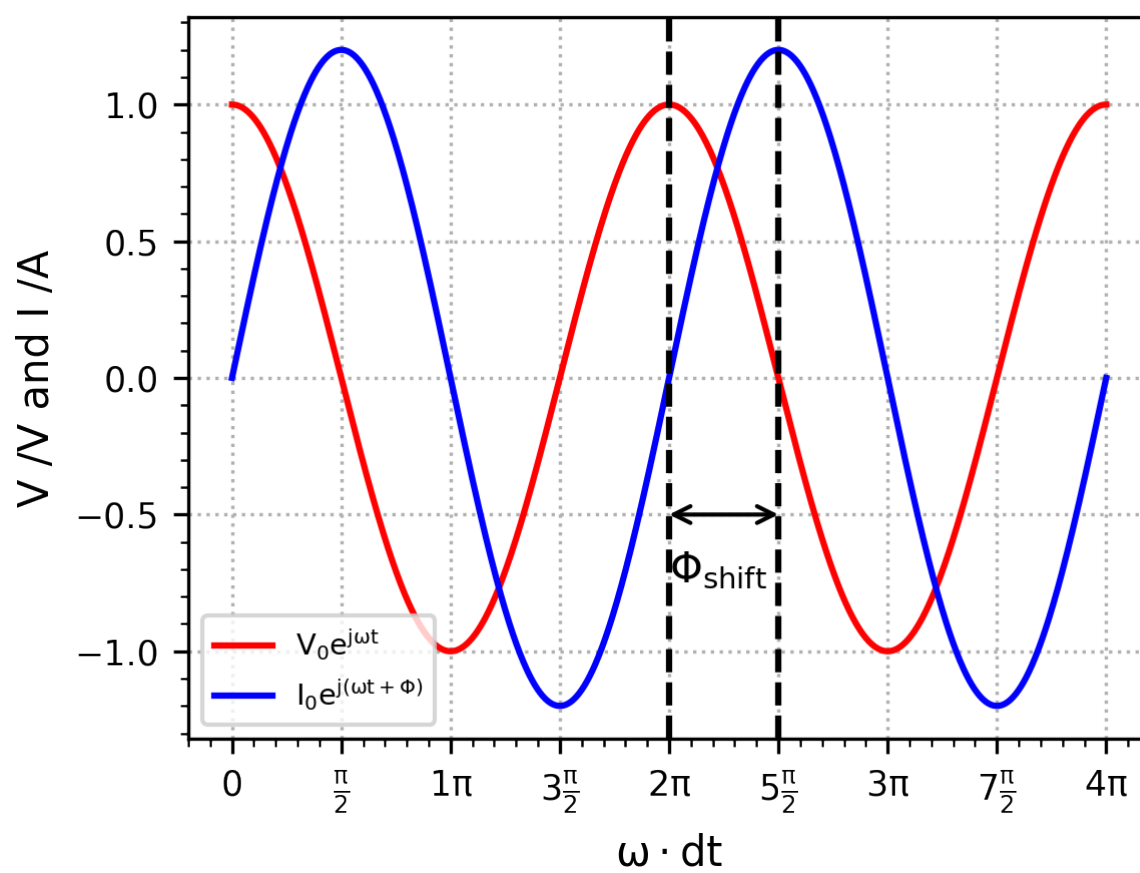


Fig. 2.2.1: EIS AC Waves

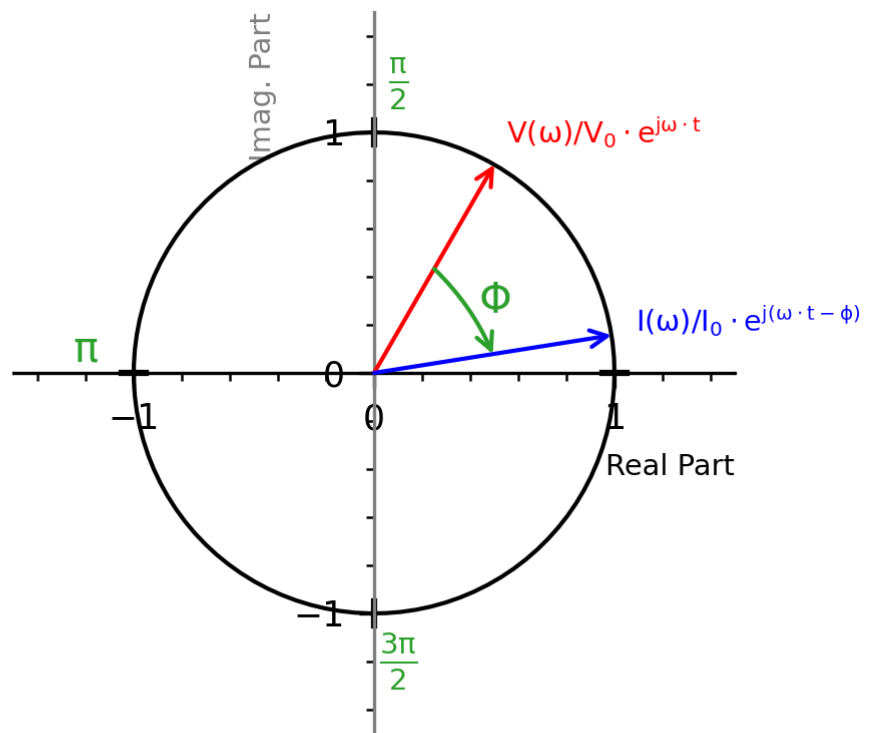


Fig. 2.2.2: Trigonometric Circle

### 2.2.3 What is EIS?

The impedance is determined from the imposed voltage/current and the measured current/voltage through the Ohm's law:

$$Z(\omega) = \frac{V(\omega)}{I(\omega)} = \frac{V_0}{I_0} e^{j\phi} = Z_0 e^{j\phi}$$

Therefore: \* Resistive behavior:  $ReZ = Z_0 \cdot \cos \phi$  \* Capacitive/Inductive behavior  $ImZ = Z_0 \cdot \sin \phi$

Sometimes, the complex admittance can also be used which is defined as the inverse of the complex impedance

$$Y(\omega) = \frac{1}{Z(\omega)}$$

### 2.2.4 Representation

The impedance  $Z(\omega)$  can be represented in two different ways as shown in:

1. **Bode plot:** shows the phase shift and magnitude changes in the applied frequency ranges as shown in [Fig. 2.2.4](#) and [Fig. 2.2.5](#)
2. **Nyquist plot:** represents the real and imaginary parts of  $Z(\omega)$  using cartesian coordinates as shown in [Fig. 2.2.3](#)

The Bode plot has great advantages for observing phase margins in which the system becomes unstable (violent phase or magnitude changes). Therefore, it is useful for the study of sensors, filters, and transistors in electronic devices.

The Nyquist plot provides insight into the possible mechanism or governing phenomena in an equivalent circuit model system. Among these two types of representations, the Nyquist plot is more often used to analyze the characteristics of electrochemical processes.

### 2.2.5 Series and Parallel Connections

- Series connection:  $Z_1 - Z_2 - \dots - Z_n = Z_{\{eq\}} = \sum Z_i$
- Parallel connection:  $Z_1/Z_2/\dots/Z_n = Z_{\{eq\}} = \left( \sum \frac{1}{Z_i} \right)^{-1}$

### 2.2.6 Equivalent Circuit Models

- The circuit model for EIS consists of a combination of electrical circuit elements:
  - ideal elements: resistors (R), capacitors (C) and inductors (L)
  - nonideal capacitor-like element: Constant Phase Element (CPE or  $Q$ )
  - diffusion elements: semi-infinite Warburg ( $W$ ), Finite Length Warburg ( $W_\delta$  or  $O$ ) and Finite Space Warburg ( $W_m$  or  $T$ )
- The circuit model represents the entire system of the electrochemical cell and therefore the aim is to construct an optimal circuit model that is physically meaningful and minimizes the number of variables.

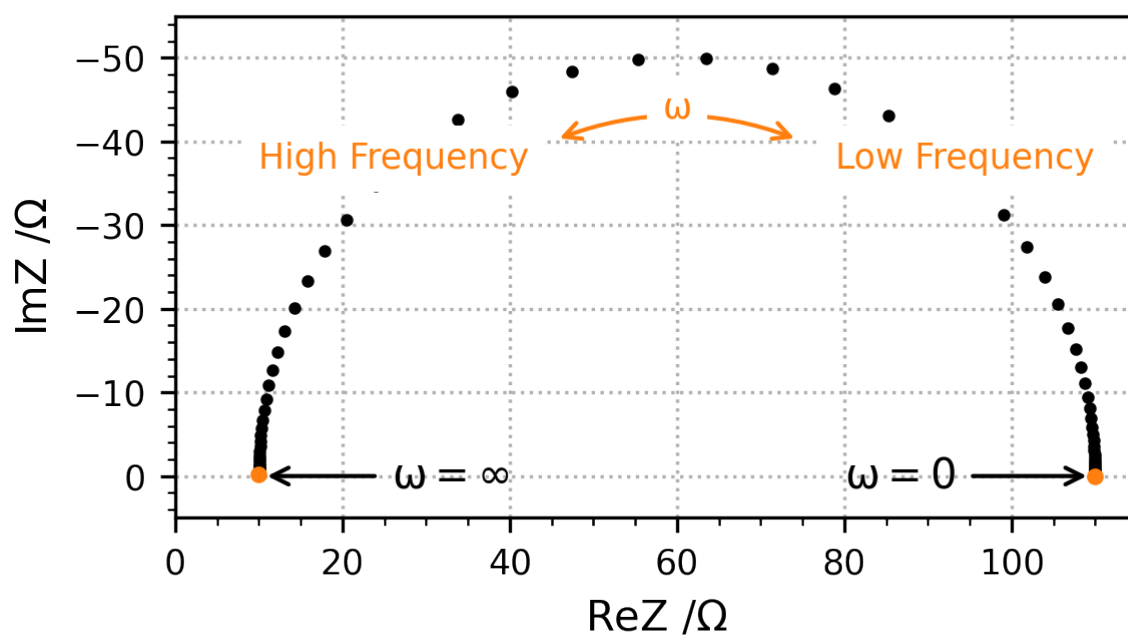


Fig. 2.2.3: Nyquist Representation

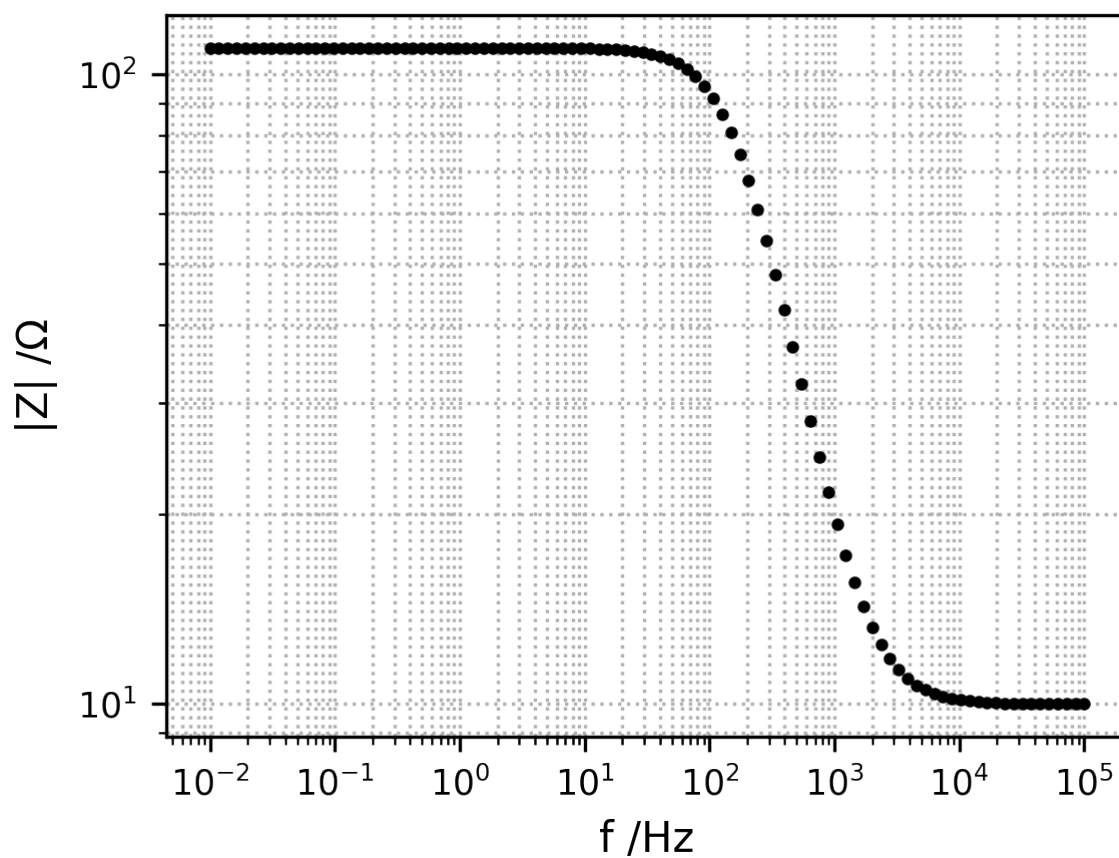


Fig. 2.2.4: Bode Modulus Representation

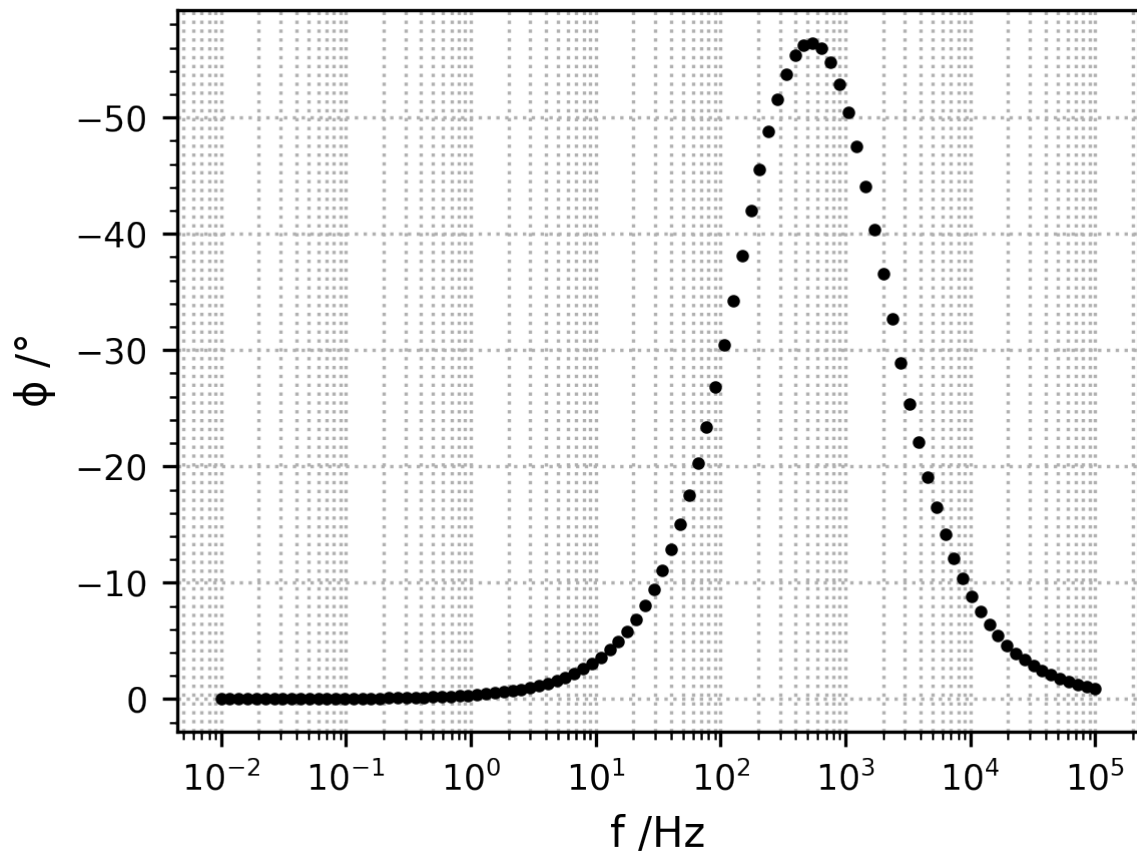


Fig. 2.2.5: Bode Phase Representation

## 2.2.7 Circuit elements

The different circuit elements available with their string representation are listed here and their Nyquist representation is shown in Fig. 2.2.6. In order to be recognized by the string parser each element must start with one or two letters defined below and can be followed by a name. The measurement model element needs an additional parameter which is the number of Voigt elements defined after an underscore.

- R[name]:  $Z(\omega) = R$
- C[name]:  $Z(\omega) = \frac{1}{jC\omega}$
- L[name]:  $Z(\omega) = jL\omega$
- W[name]:  $Z(\omega) = \frac{\sigma}{\sqrt{\omega}} \cdot (1 - j)$
- Wd[name]:  $Z(\omega) = \frac{R_s \cdot \tanh(\sqrt{j\omega\tau})}{\sqrt{j\omega\tau}}$
- Wm[name]:  $Z(\omega) = \frac{R_m \cdot \coth(\sqrt{j\omega\tau})}{\sqrt{j\omega\tau}}$
- Q[name]:  $\frac{1}{Q(j\omega)^\alpha}$
- M[name]\_n:  $Z(\omega) = R_0 + \sum_{k=0}^n \frac{R_k}{1 + jR_k C_k \omega}$
- G[name]:  $Z_G(\omega) = G \cdot (K_g + i\omega)^{-n_g}$

Inductor and Finite Space Warburg are rarely encountered in corrosion studies.

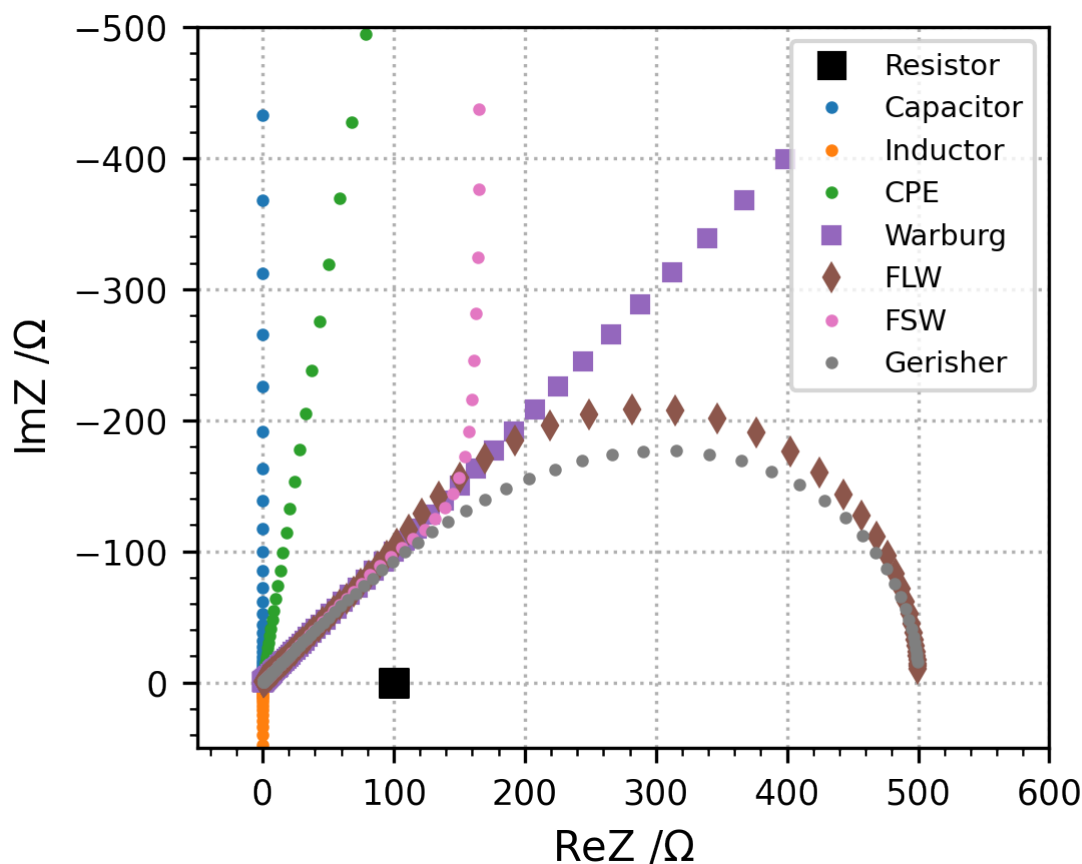


Fig. 2.2.6: Circuit Elements



## 2.2.8 Link between circuit elements and physical parameters

- Resistors can be linked to resistivity or kinetics:

$$- R = \frac{\rho \cdot d}{A}$$

$$- R = \frac{RT}{F A j_0 (\alpha_a + \alpha_c)} = \frac{RT}{A F^2 k^0 K_c (\alpha_a + \alpha_c)}$$

- Capacitors can be linked to layer thickness

$$- C = \frac{\epsilon \epsilon_0 A}{d}$$

- FS/FL Warburg element can be linked to diffusion coefficient and layer thickness:

$$- R = \frac{RT}{A F^2 \sqrt{2}} \cdot \frac{d}{D \cdot C^*}$$

$$- \tau = \frac{d^2}{D}$$

$$- \sigma = \frac{R}{\sqrt{2\tau}}$$

where,

- $R$ : resistance [ $\Omega$ ]
- $\rho$ : resistivity [ $\Omega \cdot m$ ]
- $d$ : thickness [ $m$ ]
- $A$ : Area [ $m^2$ ]
- $j_0$ : exchange current density [ $A \cdot m^{-2}$ ]
- $k^0$ : kinetics constant [ $m \cdot s^{-1}$ ]
- $K_c$ : concentration factor [ $mol \cdot m^{-3}$ ]
- $\alpha_a$ : anodic transfer coefficient
- $\alpha_c$ : cathodic transfer coefficient
- $C^*$ : bulk concentration of the diffusing species [ $mol \cdot m^{-3}$ ]

## 2.2.9 Simplified Randles Circuit

Reflects electrochemical reaction controlled only by kinetics as shown in [Fig. 2.2.7](#)

- $R_{el} + R_{ct}/C_{dl}$
- $R_{el}$ : electrolyte resistance
- $R_{ct}$ : charge transfer resistance
- $C_{dl}$ : double layer capacitance

## 2.2.10 Randles Circuit

Reflects electrochemical reaction controlled by kinetics and diffusion as shown in [Fig. 2.2.8](#)

- $R_{el} + R_{ct}/C_{dl}$
- $R_{el}$ : electrolyte resistance
- $R_{ct}$ : charge transfer resistance
- $C_{dl}$ : double layer capacitance
- $W$ : semi-infinite diffusion

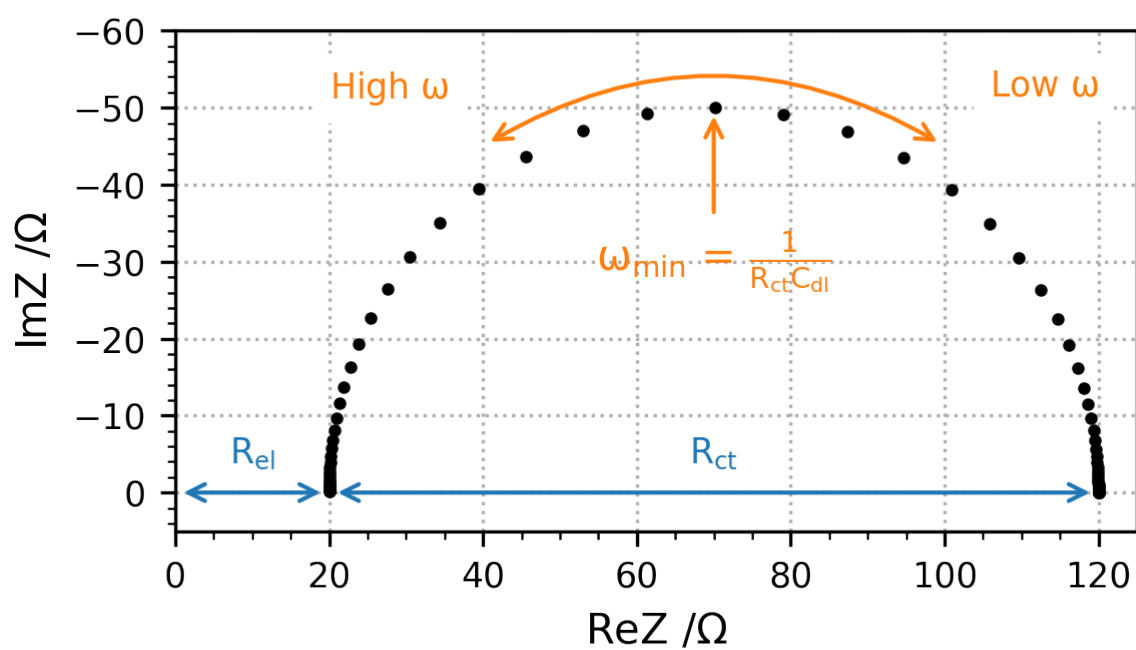


Fig. 2.2.7: Simplified Randles

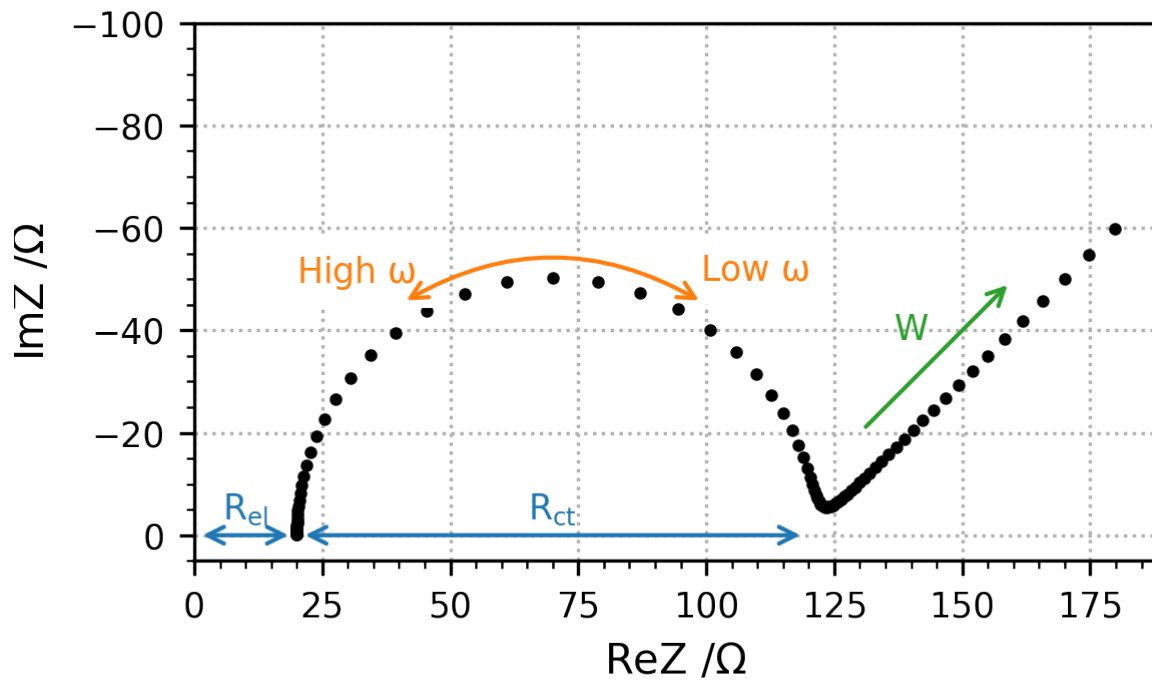


Fig. 2.2.8: Randles

### 2.2.11 Differential Impedance analysis

The differential Impedance Analysis (DIA) is based on the use of a Local Operator Model (LOM) which is a equivalent circuit for a simple Faradic reaction but has direct meaning with the experimental spectrum that is being analyzed.

The LOM operator corresponds to the equivalent circuit Rads-(R/C).

The procedure of the structural and parametric identification can be described by the following steps:

- scanning with the LOM throughout the whole frequency range with a scanning window of a single frequency
- parametric identification of the LOM parameters at every working frequency
- Frequency analysis of the LOM parameters' estimates

#### Scanning with the LOM operator

The impedance of the LOM operator is defined as shown in Eq.2.2.1:

$$Z_{LOM} = R_{ads} + \frac{R}{1 + T^2\omega^2} - j \frac{\omega RT}{1 + T^2\omega^2} \quad (2.2.1)$$

#### Parametric identification of the LOM parameters

The objective is to identify the LOM parameters  $P_j = R_{ads}, R, C, T$ .

First the effective resistance and the effective inductance are expressed:

$$R_{eff} = ReZ = R_{ads} + \frac{R}{1 + T^2\omega^2}$$
$$L_{eff} = -ImZ/\omega = \frac{RT}{1 + T^2\omega^2}$$

Derivatives of the effective resistance and inductance are:

$$\frac{dR_{eff}}{d\omega} = -R \frac{2\omega T^2}{(1 + T^2\omega^2)^2}$$
$$ImZ = -L_{eff} \cdot \omega$$
$$\frac{dImZ}{d\omega} = -\frac{ImZ}{dL_{eff}} \frac{dL_{eff}}{d\omega} = -\omega \frac{dL_{eff}}{d\omega}$$
$$\frac{dL_{eff}}{d\omega} = -RT \frac{2T^2\omega}{(1 + T^2\omega^2)^2} = -\frac{dImZ}{d\omega} \frac{1}{\omega}$$

Expression of the LOM parameters  $P_i$ :

$$T(\omega) = \frac{\frac{dL_{eff}}{d\omega}}{\frac{dR_{eff}}{d\omega}} = \frac{dL_{eff}}{dR_{eff}}$$
$$R(\omega) = -\frac{dR_{eff}}{d\omega} \cdot \frac{(1 + T^2\omega^2)^2}{2\omega T^2}$$
$$R_{ads}(\omega) = R_{eff}(\omega) - \frac{R}{1 + T^2\omega^2}$$
$$C(\omega) = \frac{T}{R}$$

## Temporal analysis

The temporal analysis computes the logarithmic values of the LOM parameters  $L_j = a, r, c, t$  with respect to  $\nu$  as defined in Eq.2.2.2:

$$\begin{aligned} L_j &= \log_{10} P_j = \log_{10} Rads, \log_{10} R, \log_{10} C, \log_{10} T \\ L_j &= a, r, c, t \\ \nu &= \log_{10} \frac{1}{\omega} \end{aligned} \quad (2.2.2)$$

## Differential temporal analysis

The differential temporal analysis computes the derivatives  $d_j$  of  $L_j$  with respect to  $\nu$  as defined in Eq.2.2.3

$$d_j = \frac{dL_j}{d\nu} = da, dr, dc, dt \quad (2.2.3)$$

## Spectral analysis

The spectral analysis is obtained by accumulating frequency bands with approximatively equal values of the parameters  $L_j$ . The amplitude of the individual spectral line  $S_{j,l}$  can be expressed as shown in Eq.2.2.4.

$$\begin{aligned} S_{j,l} &= \sum_1^N B(L_{j,i}) \\ B(L_{j,i}) &= w_0 \text{ if } l < L_{j,l} < l + s \\ B(L_{j,i}) &= 0 \text{ otherwise} \\ w_0 &= N_{frequencies} / N_{decades} \end{aligned} \quad (2.2.4)$$

The spectral line is expressed in dB.

## Differential spectral analysis

The differential spectral analysis is obtained by accumulating frequency bands with approximatively equal values of the parameters  $d_j$ . The amplitude of the individual spectral line  $S_{j,l}$  can be expressed as shown in Eq.2.2.5.

$$\begin{aligned} S_{j,l} &= \sum_1^N B(d_{j,i}) \\ B(d_{j,i}) &= w_0 \text{ if } l < d_{j,l} < l + s \\ B(d_{j,i}) &= 0 \text{ otherwise} \\ w_0 &= N_{frequencies} / N_{decades} \end{aligned} \quad (2.2.5)$$

The spectral line is expressed in dB.

An example with a simple RC circuit:

## 2.3 PEC

### 2.3.1 Introduction

PEC takes advantage of the photovoltaic effect, discovered by [6] in 1839, that occurs at the interface of a semiconductor and an electrolyte. In fact, the first experience showed the occurrence of a photopotential and a photocurrent under illumination when a silver electrode, covered with an oxide layer, was immersed in an acidic medium and

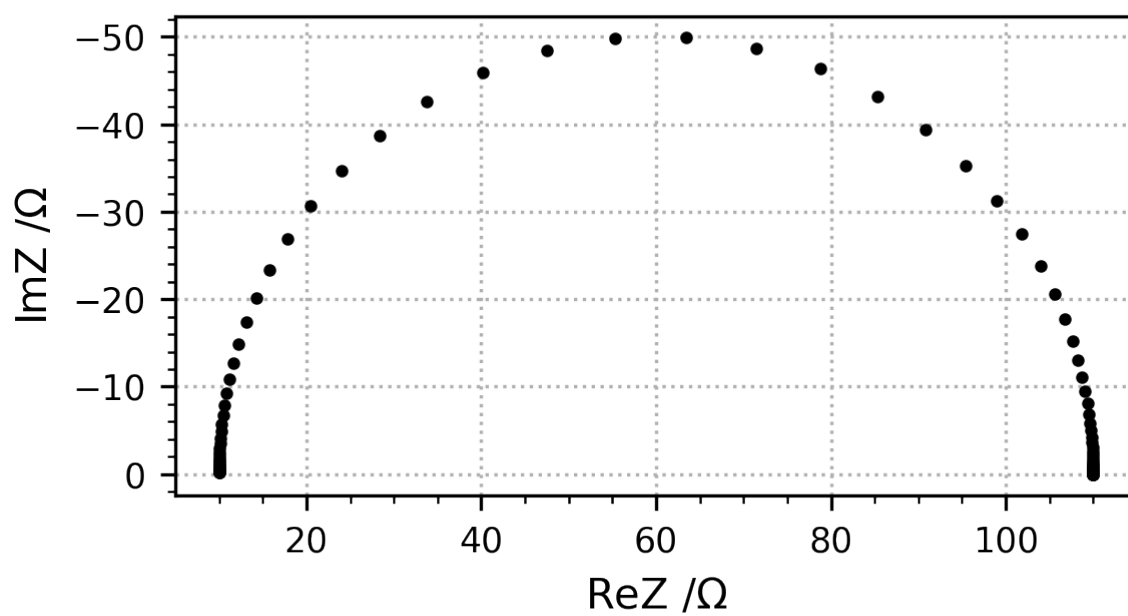


Fig. 2.2.9: Simple RC

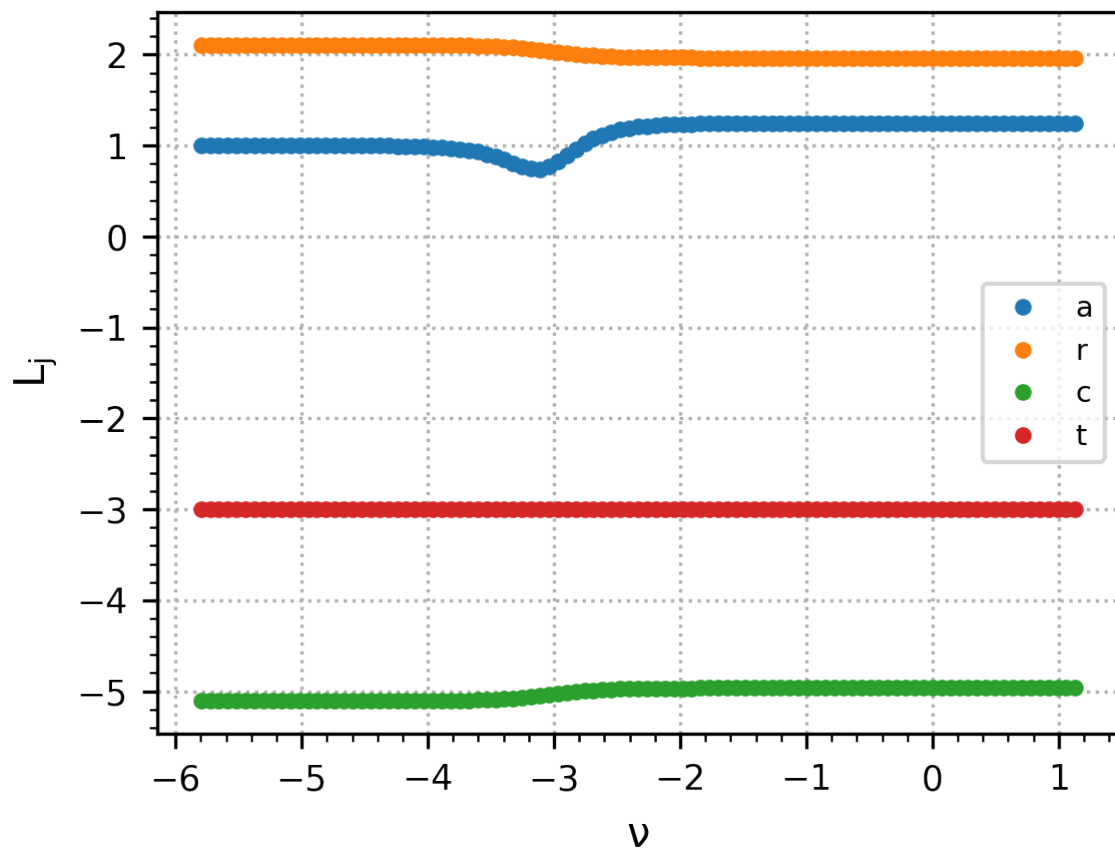


Fig. 2.2.10: Temporal Analysis

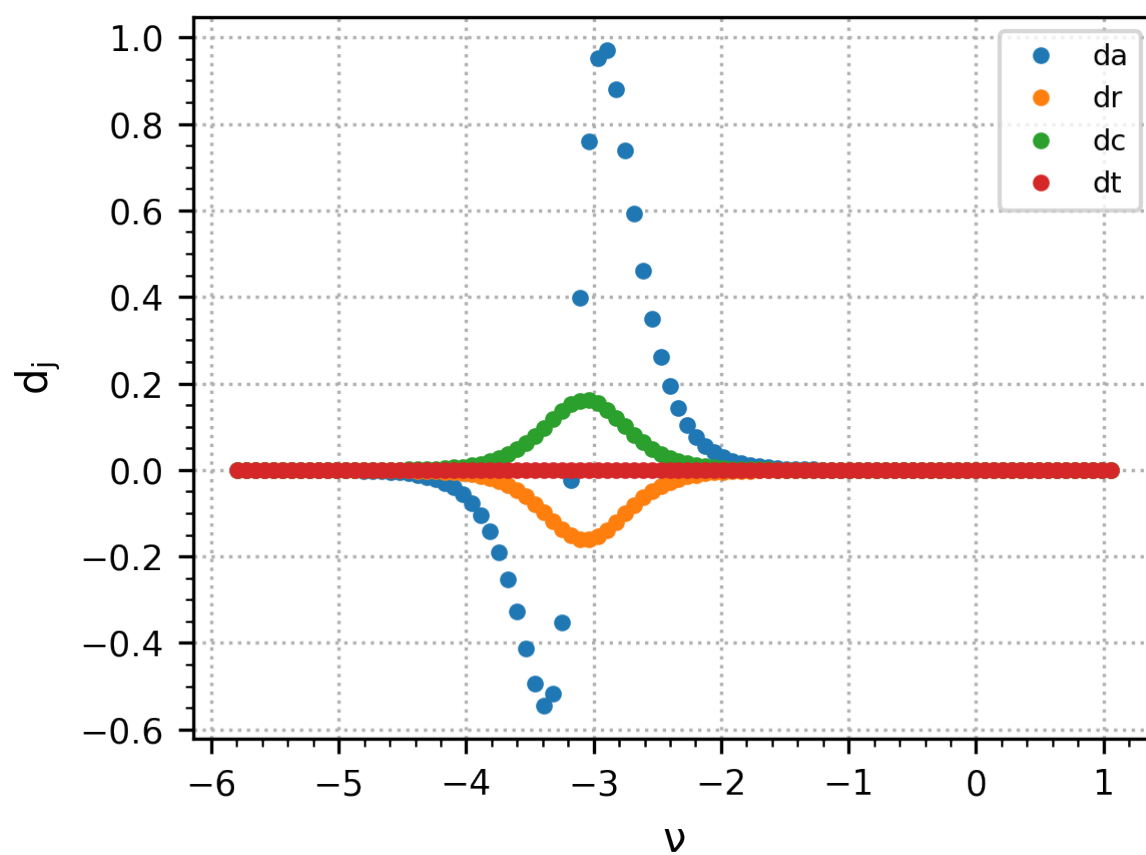


Fig. 2.2.11: Differential Temporal Analysis



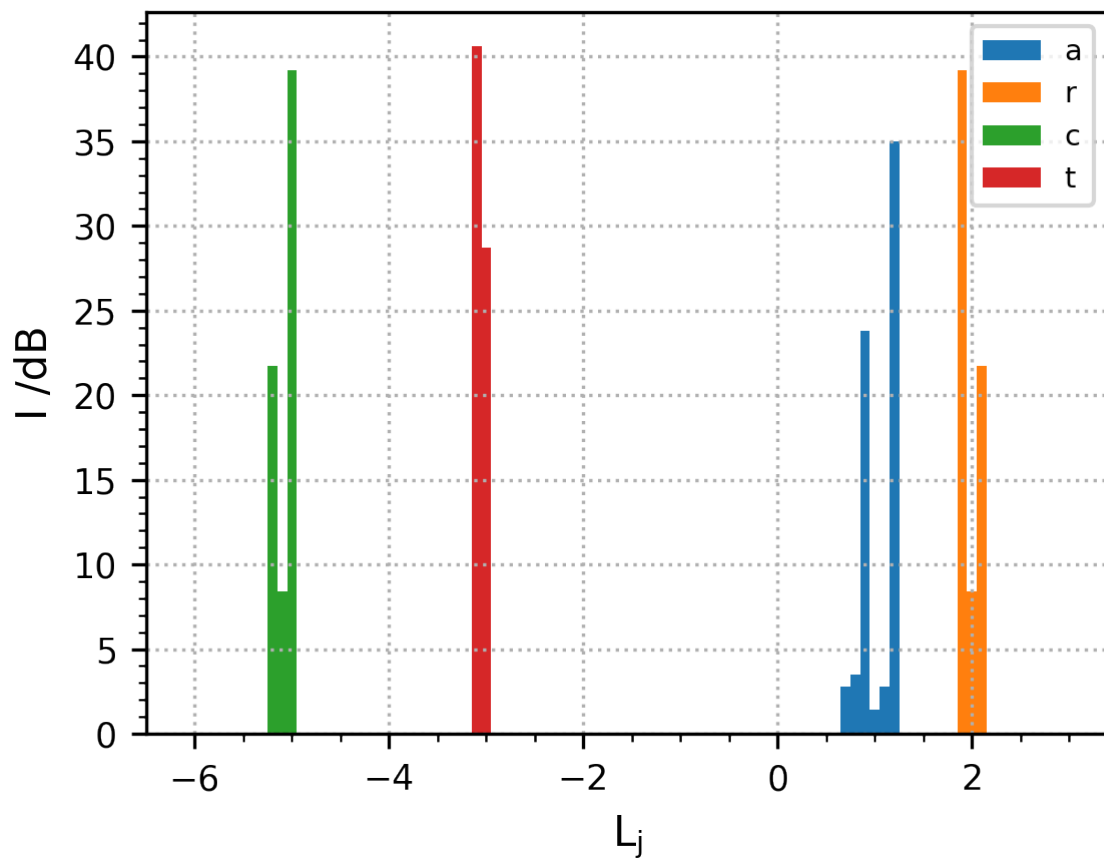


Fig. 2.2.12: Spectral Analysis

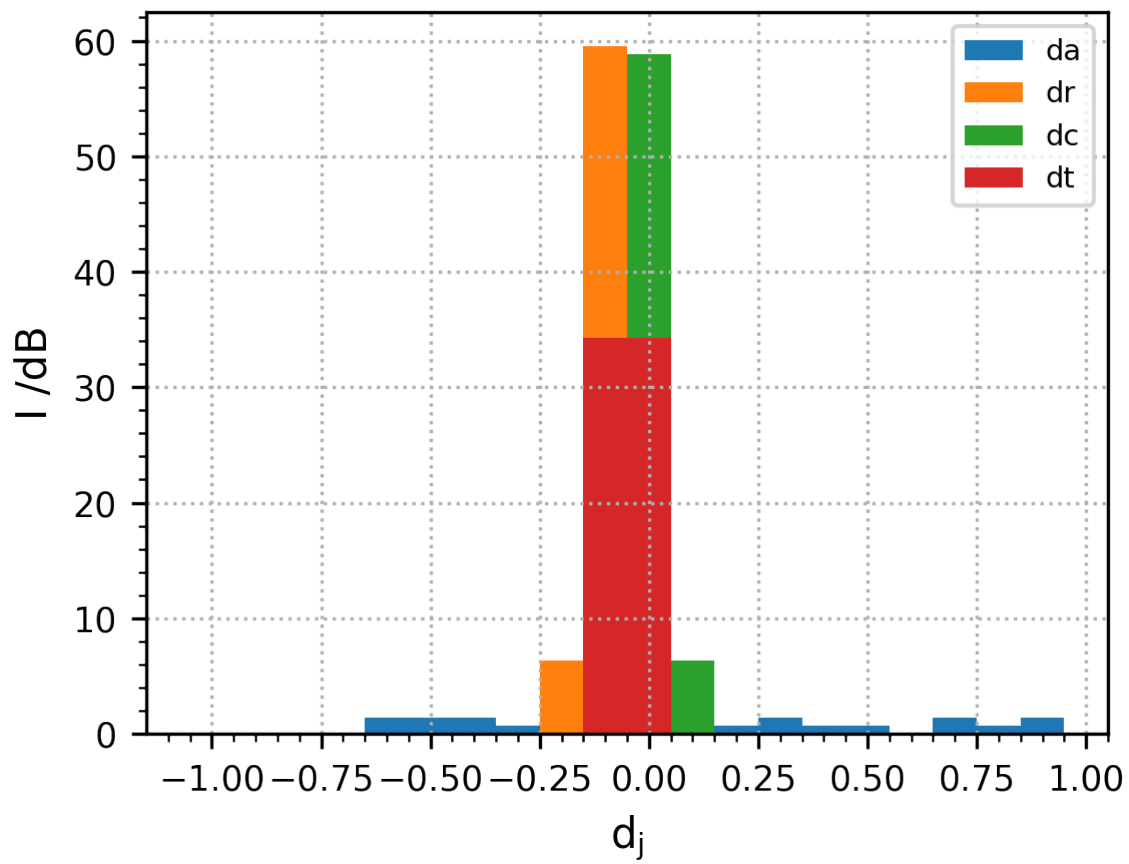


Fig. 2.2.13: Differential Spectral Analysis

connected to a platinum electrode. Nonetheless, the first studies focused on the understanding of the interfacial processes were performed much later [7, 8, 9].

The basics of photoelectrochemistry and application examples are presented here and they are largely described in the literature [10, 11, 12, 13, 14, 15]. Several hypotheses are needed in order to apply the theoretical concepts:

- semiconductors are considered to be ideal i.e. crystalized and homogeneous
- the dielectric constant of the semiconductor is independent of the light wavelength
- the capacity of the Helmholtz layer is greater than the capacity of the space charge capacitance
- the potential drop in the Helmholtz layer is independent of the applied potential and is negligible

The hypotheses are rarely fully respected in the case of oxides or passive films formed on common alloys. Nonetheless, the literature shows that the developed models can be applied to non-ideal systems such as oxides or passive layers.

Solids are generally classified into three groups: *conductors*, *semiconductors* and *insulators*. Each category can be illustrated with a specific band structure as shown in figure Fig. 2.3.1 [16]. Valence and conduction bands correspond to the allowed energy states for the electrons. The lowest energy level of the conduction band is labeled  $E_c$  and the highest energy level of the valence is labeled  $E_v$ . They are separated by a band gap,  $E_g$ , with no allowed energy states. The repartition of the electrons among both bands are described by the position of the Fermi level,  $E_F$ , which represents the highest energy state that can be occupied at 0K.

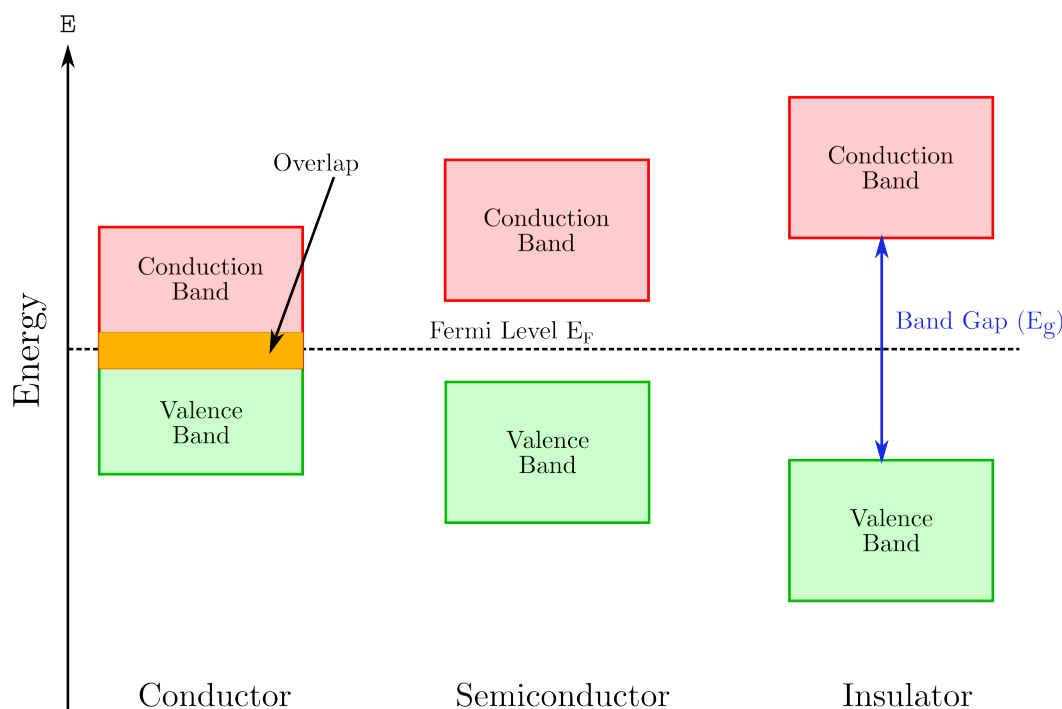


Fig. 2.3.1: Band Structure

The electronic conduction is due to the movement either of the negatively charge electrons in the conduction band or the positively charged holes in the valence band or both simultaneously. Consequently, the conduction depends on the number of available charge carriers in the conduction and valence bands. In conductors, an overlap of the conduction and the valence bands occurs which means that the highest allowed energy band is partially filled. The distinction between a semiconductor and an insulator is less obvious because the conduction depends on the band

gap and the energy provided by the environment to the electron from the valence band in order to jump into the conduction band.

In semiconductors, charge carriers can be generated by three mechanisms: *thermal*, *excitation*, *photoexcitation*, *doping*, as shown in figure Fig. 2.3.2.

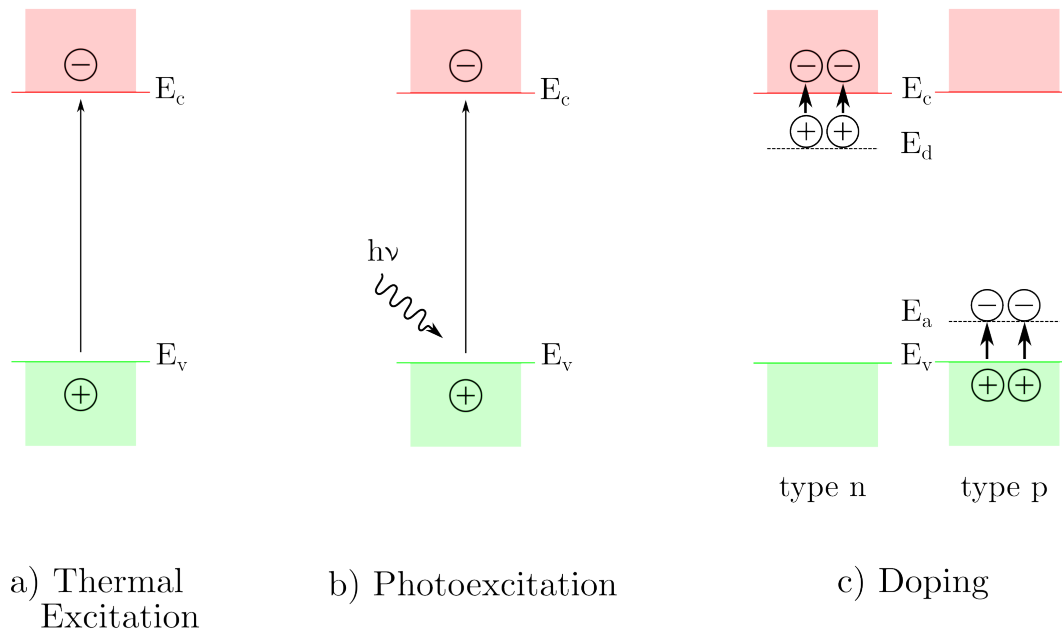


Fig. 2.3.2: Schematic representation of the mechanisms generating charge carriers in semiconductors

In the case of very low band gaps, thermal excitation can be enough to eject an electron from the valence band into the conduction band. Photoexcitation ejects electrons from the valence band into the conduction band when an incident photon, with an energy greater than the band gap, is absorbed. Doping introduces additional energy levels located in between the conduction and valence bands.

Doping occurs when the stoichiometry is altered or when impurities are introduced in the crystallographic lattice of the semiconductor. In the case of n-type semiconductors, the donor energy levels  $E_d$  lie just under the conduction band. The electrons from the donor levels are ejected by thermal excitation. Consequently, the majority charge carriers are negatively charged electrons in the conduction band. Similarly, the acceptor energy levels  $E_a$ , of p-type semiconductors, lie just above the valence band. The latter trap electrons from the valence band and therefore create holes. Consequently, the majority charge carriers are positively charged holes.

The Fermi level  $E_F$  in intrinsic semiconductors is located at the mid-gap. The n-type and p-type doping shift the Fermi level towards band edges  $E_c$  and  $E_v$ , respectively. The figure Fig. 2.3.3 shows the position of the Fermi level with respect to the semiconductor types.

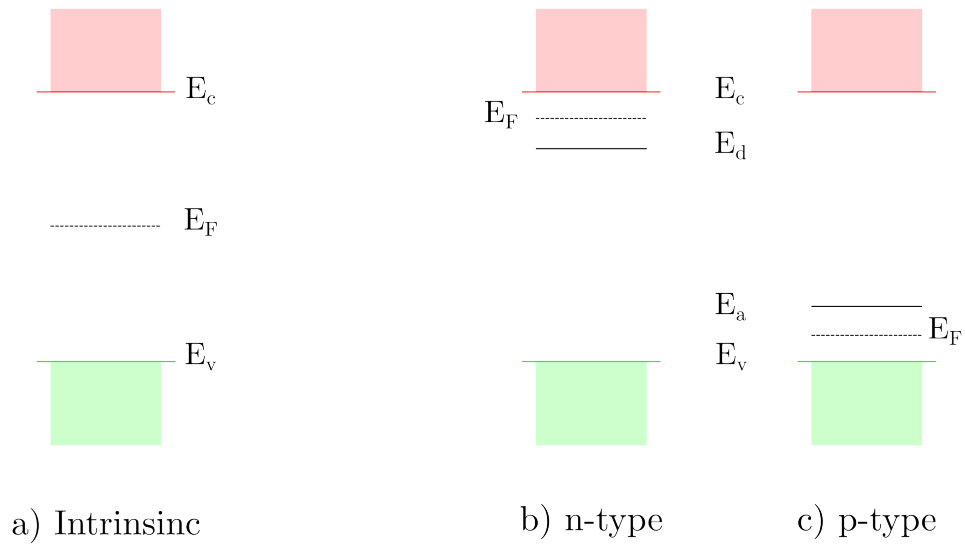


Fig. 2.3.3: Schematic representation of the Fermi level position

### 2.3.2 Semiconductor/electrolyte interface in dark

### 2.3.3 Semiconductor/electrolyte interface under illumination



## RELEASE NOTES

### 3.1 ecx 0.1.0 Release Note

#### 3.1.1 Changes

- Implementation of eis + C API
- Python wrappers for eis.
- Documentation with sphinx.

#### 3.1.2 Download

`ecx`

`pyecx`

#### 3.1.3 Contributors

Milan Skocic

#### 3.1.4 Commits

Full Changelog: <https://github.com/MilanSkocic/pyecx/compare/...0.1.0>





## AUTOGENERATED DOCUMENTATION

### 4.1 ecx

#### 4.1.1 Fortran

Fortran code API

#### 4.1.2 C API

##### Common headers

- *ecx\_types.h*

```
/**
 * @file ecx_types.h
 * @brief Type C header for th ecx library.
 * @details Compatibilty layer for handling complex numbers with MSC.
 * It is imported by all headers for each submodule.
 */
#ifndef ECX_TYPES_H
#define ECX_TYPES_H

#include <complex.h>
#if _MSC_VER
typedef _Dcomplex ecx_cdouble; /**< Cross platform complex type. */
#define ecx_cbuild(real, imag) (_Cbuild(real, imag)) /**< Cross platform complex type.
↳ constructor*/
#else
typedef double _Complex ecx_cdouble; /**< Cross platform complex type. */
#define ecx_cbuild(real, imag) (real+I*imag) /**< Cross platform complex type.
↳ constructor*/
#endif
#endif
```

- *ecx.h*

```
/**
 * @file ecx.h
 * @brief Main C header for th ecx library.
 * @details Includes all the headers for each submodule.
 */
#ifndef ECX_H
#define ECX_H
```

(continues on next page)

(continued from previous page)

```
#include "ecx_core.h"
#include "ecx_eis_capi.h"
#include "ecx_pec_capi.h"
#include "ecx_kinetics.h"

#endif
```

## EIS headers

```
/**
 * @file ecx_eis_capi.h
 * @brief EIS C header for th ecx library.
 * @details Complex impedance
 */
#ifndef ECX_EIS_CAPI_H
#define ECX_EIS_CAPI_H
#include "ecx_types.h"
#include "ecx_core.h"

extern void ecx_capi_zr(double *w, double R, size_t n, ecx_cdouble *Z);
extern void ecx_capi_zc(double *w, double C, size_t n, ecx_cdouble *Z);
extern void ecx_capi_zl(double *w, double L, size_t n, ecx_cdouble *Z);
extern void ecx_capi_zcpe(double *w, double Q, double a, size_t n, ecx_cdouble *Z);
extern void ecx_capi_zw(double *w, double s, size_t n, ecx_cdouble *Z);
extern void ecx_eis_capi_z(double *p, double *w, ecx_cdouble *z, char e, size_t k,
↪size_t n, int *errstat);

#endif
```

## PEC headers

```
/**
 * @file ecx_pec_capi.h
 * @brief EIS C header for th ecx library.
 */
#ifndef ECX_PEC_CAPI_H
#define ECX_PEC_CAPI_H
#include "ecx_types.h"
#include "ecx_core.h"

#endif
```

## 4.2 pyipaws

C extension wrapping the EIS module of the Fortran ecx library.

`pyecx.eis.z()`

`z(e, w, p)→memview`

Compute the complex impedance of the element `e`.



## BIBLIOGRAPHY

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