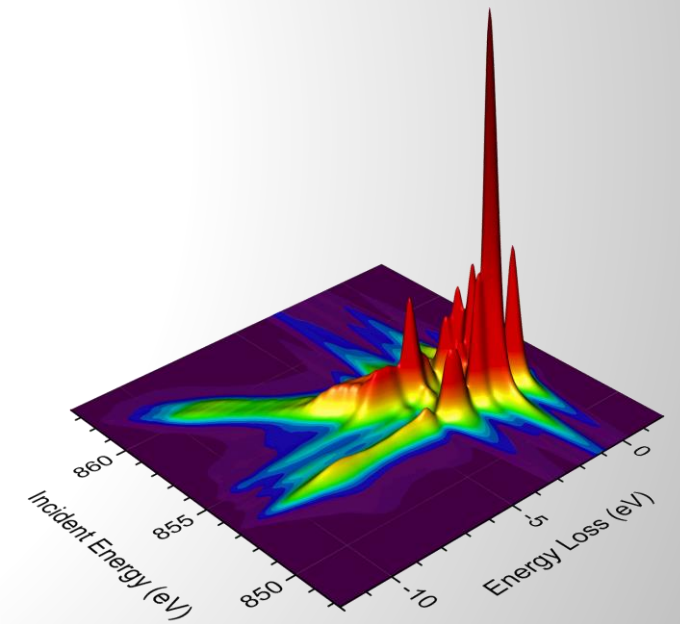


Analysis of soft XAS and RIXS with multiplet models using Quanty

Part 1: Understanding the Spectra

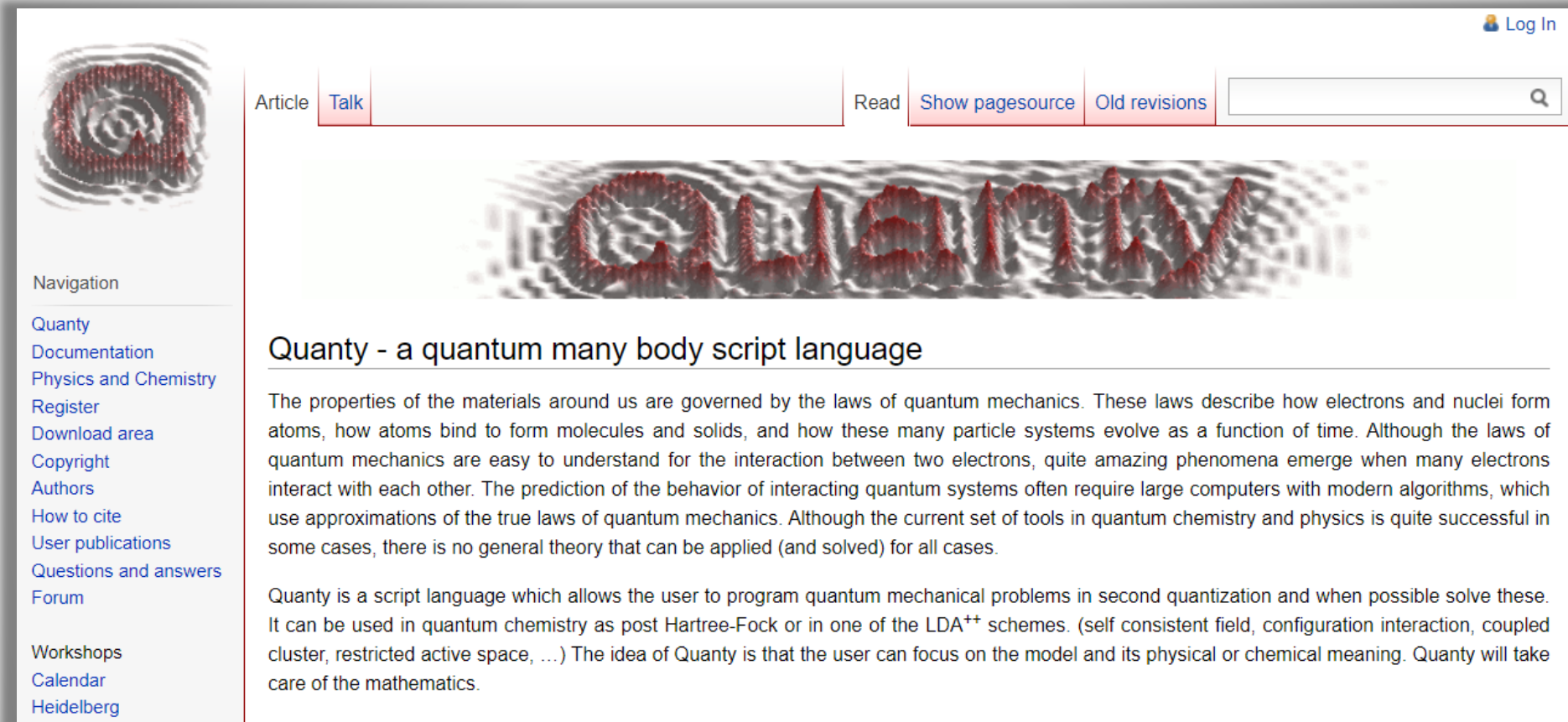
Part 2: Quanty Computations



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June 20, 2023

https://quandy.org



The screenshot shows the Quandy website interface. On the left is a navigation menu with links: Quandy, Documentation, Physics and Chemistry, Register, Download area, Copyright, Authors, How to cite, User publications, Questions and answers, Forum, Workshops, Calendar, and Heidelberg. The main content area features a large 3D visualization of a quantum system, possibly a molecule or a lattice structure, rendered in red and white. Below this is the article title 'Quandy - a quantum many body script language' and two paragraphs of text. The top navigation bar includes 'Article', 'Talk', 'Read', 'Show pagesource', 'Old revisions', and a search box. A 'Log In' button is visible in the top right corner.

Navigation

- [Quandy](#)
- [Documentation](#)
- [Physics and Chemistry](#)
- [Register](#)
- [Download area](#)
- [Copyright](#)
- [Authors](#)
- [How to cite](#)
- [User publications](#)
- [Questions and answers](#)
- [Forum](#)
- [Workshops](#)
- [Calendar](#)
- [Heidelberg](#)

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Quandy - a quantum many body script language

The properties of the materials around us are governed by the laws of quantum mechanics. These laws describe how electrons and nuclei form atoms, how atoms bind to form molecules and solids, and how these many particle systems evolve as a function of time. Although the laws of quantum mechanics are easy to understand for the interaction between two electrons, quite amazing phenomena emerge when many electrons interact with each other. The prediction of the behavior of interacting quantum systems often require large computers with modern algorithms, which use approximations of the true laws of quantum mechanics. Although the current set of tools in quantum chemistry and physics is quite successful in some cases, there is no general theory that can be applied (and solved) for all cases.

Quandy is a script language which allows the user to program quantum mechanical problems in second quantization and when possible solve these. It can be used in quantum chemistry as post Hartree-Fock or in one of the LDA⁺⁺ schemes. (self consistent field, configuration interaction, coupled cluster, restricted active space, ...) The idea of Quandy is that the user can focus on the model and its physical or chemical meaning. Quandy will take care of the mathematics.

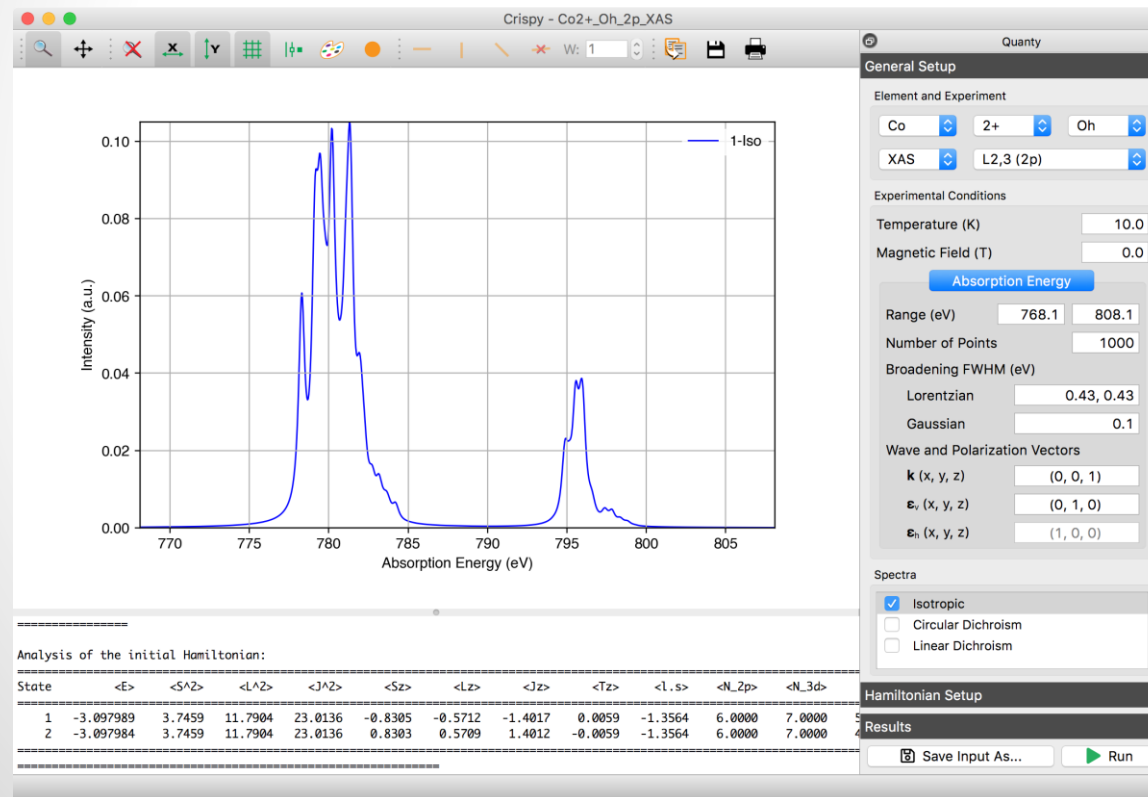
Lots of information on the website

– installation, execution, tutorials/examples, and full documentation of all capabilities

Quanty is designed as an executable program that reads input files written in the Lua scripting language. This makes it extremely flexible and powerful.

However, a 3rd party graphical user interface called Crispy has been written for Quanty, which works well for many standard types of calculations

<https://www.esrf.fr/computing/scientific/crispy/index.html>



A Typical Quantity Computation

Define a Hamiltonian (H) operator to model your system

```
H = F0dd*OppF0_3d + F2dd*OppF2_3d + F4dd*OppF4_3d
```

The majority of the work

Compute some eigenstates of this H

```
psiList = Eigensystem(H, StartRestrictions, Npsi)
```

Compute properties for the eigenstates of this H

```
E0 = psiList[1]*H*psiList[1]
```

$$E_0 = \langle \Psi_0 | H | \Psi_0 \rangle$$

Compute XAS spectra for this H

```
XASSpectra = CreateSpectra(XASH, T2p3dx, psiList[1], { ...
```

Compute RIXS spectra for this H

```
RIXSSpectra = CreateResonantSpectra(XASH, H, T2p3dx, T3d2py, psiList[1], { ...
```

Calculations of NiO with Increasing Complexity

NiO is a good example, as simple models like **crystal field theory** do well, but we can also see definite improvements when going all the way to high complexity **impurity models**

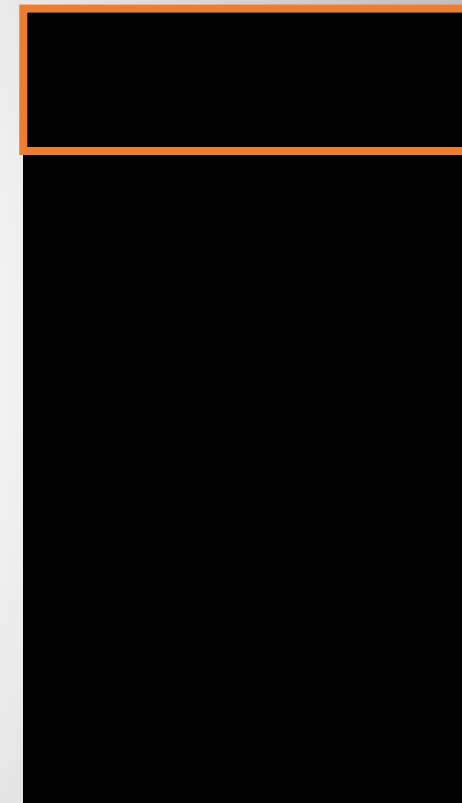
In this example, we'll learn how to compute NiO XAS and RIXS for three of the models discussed in Part 1:

- atomic model
- crystal field model
- ligand field model

Anatomy of a Typical Quany File

Define orbital indices:

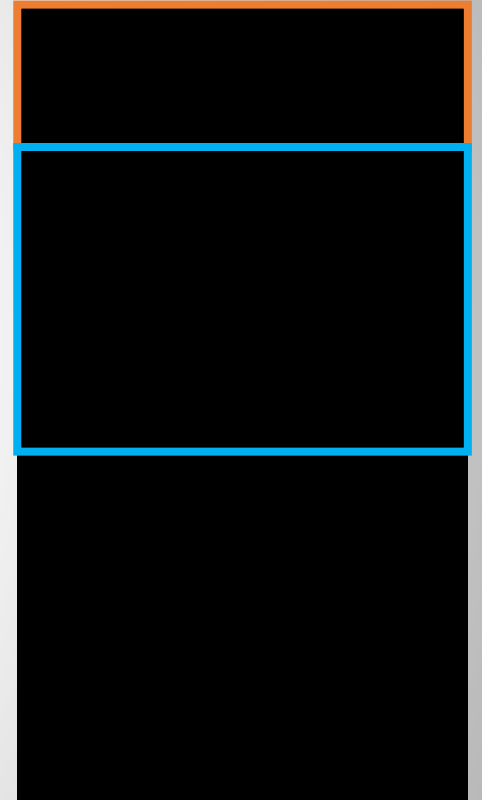
```
NF=28  
NB=0  
IndexDn_2p={ 0, 2, 4}  
IndexUp_2p={ 1, 3, 5}  
IndexDn_3s={ 6}  
IndexUp_3s={ 7}  
IndexDn_3d={ 8,10,12,14,16}  
IndexUp_3d={ 9,11,13,15,17}  
IndexDn_Ld={18,20,22,24,26}  
IndexUp_Ld={19,21,23,25,27}
```



Anatomy of a Typical Quany File

Define operators:

```
Akm = PotentialExpandedOnClm("Oh", 2, {0.6,-0.4})  
Akm = PotentialExpandedOnClm("Oh", 2, {1,0})  
Akm = PotentialExpandedOnClm("Oh", 2, {0,1})  
  
OpptenDq_3d = NewOperator("CF", NF, IndexUp_3d, IndexDn_3d, Akm)  
OppNeg_3d    = NewOperator("CF", NF, IndexUp_3d, IndexDn_3d, Akm)  
OppNt2g_3d  = NewOperator("CF", NF, IndexUp_3d, IndexDn_3d, Akm)
```



Anatomy of a Typical Quany File

Define scalar parameters:

```
nd = 8

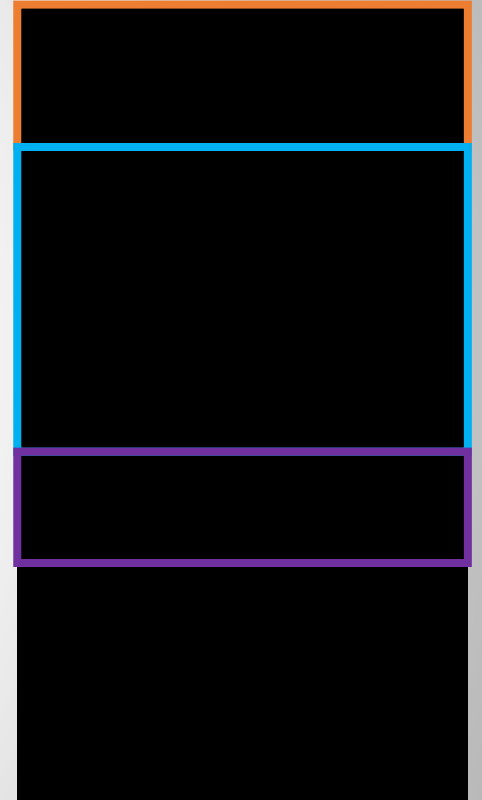
Udd    = 7.3
Upd    = 8.5
Delta  = 4.7

F2dd   = 11.14
F4dd   = 6.87
F2pd   = 6.67
G1pd   = 4.92
G3pd   = 2.80

tenDq  = 0.56

Veg    = 2.06
Vt2g   = 1.21

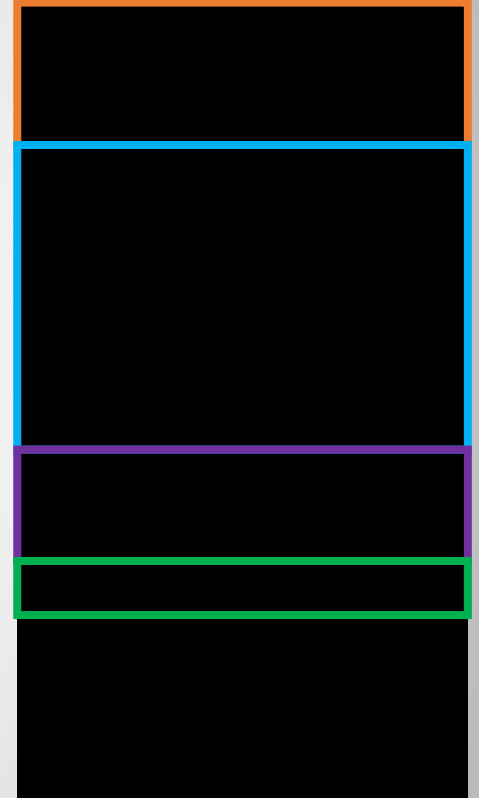
zeta_3d = 0.081
zeta_2p = 11.51
```



Anatomy of a Typical Quany File

Construct Hamiltonian:

```
H = F0dd*OppF0_3d +  
    F2dd*OppF2_3d +  
    F4dd*OppF4_3d +  
    zeta_3d*Oppldots_3d +  
    Bz*(2*OppSz_3d + OppLz_3d) +  
    tenDq*OpptenDq_3d +  
    tenDqL*OpptenDq_Ld +  
    Veg * OppVeg +  
    Vt2g * OppVt2g +  
    ed * OppN_3d +  
    eL * OppN_Ld
```

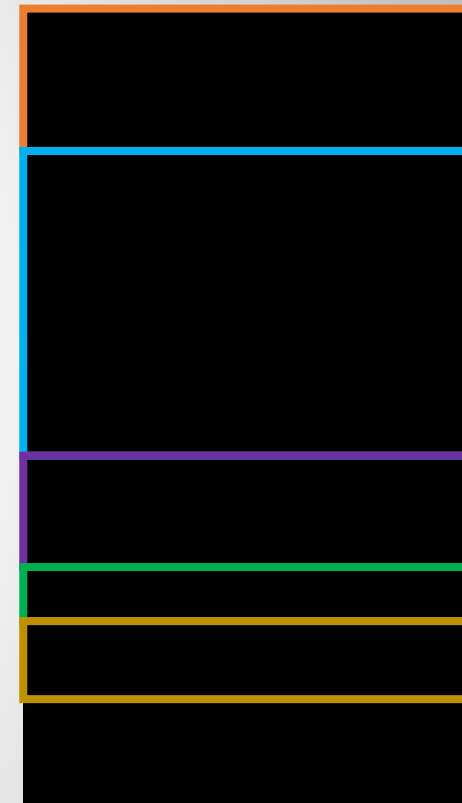


Anatomy of a Typical Quany File

Calculate ground state and expectation values:

```
psiList = Eigensystem(Hamiltonian, StartRestrictions, Npsi)

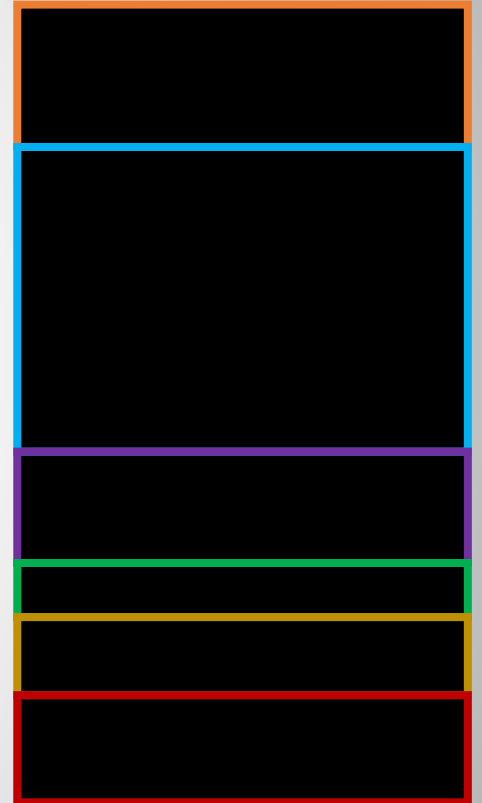
print("E0: ", psiList[1]*H*psiList[1])
print("N3d: ", psiList[1]*OppN_3d*psiList[1])
print("S^2: ", psiList[1]*OppSsqr*psiList[1])
```



Anatomy of a Typical Quanta File

Calculate spectra and save to file:

```
XASSpectra = CreateSpectra(XASHamiltonian, T2p3dx, psiList[1],  
                           {"Emin",-10}, {"Emax",20}, {"NE",3500}, {"Gamma",0.45})  
  
XASSpectra.Broaden(0,0.3)  
XASSpectra.Print({"file", "RIXSL23M45_XAS.dat"})
```



[Live Demo]

Concluding Remarks

Input files for the cases discussed today are available

Many more tutorials on Quanty website

Quanty workshops are held on an almost annual basis: <https://quany.org/workshop/start>

There is a Quanty help forum: <https://quany.org/forum/start>

Quanty is a great tool to learn about quantum mechanics (but you also don't need to know QM, for example if using Crispy)

Common pitfall: adjusting parameters to unfeasible values

- Compare charge transfer and hybridization parameters to established works on similar materials
- Are crystal field energies reasonable, given the structure of your material?
- Are your Slater integrals reasonable, given the element and oxidation state?
- Are the eigenstate properties comparable to expectations?
 - i.e. have other works determined spin state, electron occupations, etc?

Good reference: “Core Level Spectroscopy of Solids”, Frank de Groot and Akio Kotani, CRC Press (2008)