UPPSALA UNIVERITY

Bachelor thesis

Gossiping electrons

Strong decoherence from screening

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Abstract

In a strongly correlated material the localized electrons, typically the electrons in the 3d-orbitals, become entangled with each other through the Coulomb interaction. However, these electrons also interact with more mobile (itinerant) electrons in the s- and p-orbitals. The latter process called screening as it effectively reduces the strength of the interaction between the 3d-electrons. A less studied and often neglected effect of the screening is that it also entangles the 3d-electrons with the itinerant electrons, which is equivalent to a leakage of quantum information from the 3d-electrons to the environment. This process leads to decoherence since it causes the 3d-electrons to effectively lose some of their quantum mechanical properties. But what does this mean for our understanding of strongly correlated materials and can this decoherence effect be of such magnitude that neglecting it may qualitatively affect the calculated material properties? This is the question this report tries to answer, but for a minimal impurity model consisting of an atom and a few surrounding bath orbitals.

Sammanfattning

I korrelerade atomer kan lokaliserade elektroner, som elektroner i 3d orbitaler, bli kvantmekaniskt sammanflätade med varandra genom coulomb-växelverkan. Dessa elektroner kan även växelverka med mer mobila elektroner, som elektroner i s- och p-orbitaler. Denna process kallas för skärmning eftersom den effektivt sätt reducerar styrkan på repulsionen mellan elektronerna i 3d-orbitalerna. En mindre känd och ofta ignorerad effekt från skärmningen är att elektronerna i 3d-orbitalerna blir kvantmekaniskt sammanflätade med de mobila elektronerna på ett irreversibelt sätt. Detta är ekvivalent med att information om d-elektronernas position läcker ut till omgivningen. Denna informationsläcka kallas för dekoherens eftersom den leder till att d-elektronerna förlorar en del av sina kvantmekaniska egenskaper. Frågan blir således vad dekoherens kan ha för betydelse för starkt korrelerade materials egenskaper. Kan denna effekt vara av sådan magnitud att det ger oss en helt felaktig bild om den negligeras? Detta är vad denna rapport syftar till att svara på.

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Introduction

Electrons have quantum mechanical characteristics, and an implication of this is that we cannot describe these kinds of systems using ordinary classical physics, such as the superpositions of electrons. Electrons in orbitals can interact with each other through coulomb interaction, and with this interaction they can get information about one another, like position for example. Many times, one would like to study only a few electrons instead of the whole system, which would make these electrons a subsystem. But to only consider a subsystem without explicitly including the remaining electrons in the calculation, we need to approximately compensate for the effect these electrons have on the subsystem. This compensation is called screening [1].

When we make models and simulations of quantum states, it is common to only consider the coulomb interactions effect on electrons within separate subsystems [2]. However, this approach does not take into consideration the potential leakage of information between the subsystems caused by the interaction between the electrons. The question we aim to investigate is thus, how much the electron interaction contributes to this leakage of information what we call decoherence.

The purpose of this study is to understand how we can improve theoretical methods used to describe electrons in materials. And we will try to answer the question, is it necessary to create a better/more advanced description of screening that also takes decoherence into consideration?

Quantum mechanics for a beginner

To understand the problem, we need to get an understanding of some fundamental principles about the behaviors of electrons in quantum mechanical systems (QM systems).

Wave-particle duality

Electrons seemingly behave both like waves and particles. This is called the wave-particle duality which states that entities like electrons can be described as either one or the other depending on the circumstances. The most known example of this would be the double slit experiment that shows how an electron interferes with itself just like a mechanical wave would do.

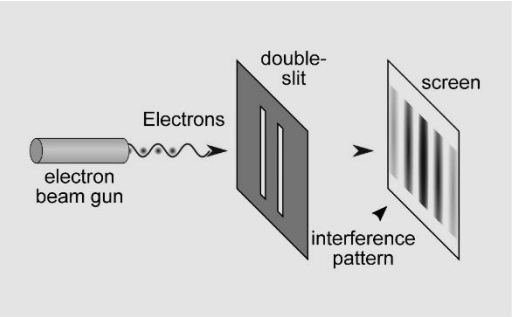

Figure 1: An illustration of an electron passing a double slit, interference with itself. Image adapted from [3]

The electron passes through both slits and interfere with itself, creating the interference pattern.

If we place detectors in both slits, we instead see the electron behave like a particle, with no interference and going through one or the other slit.

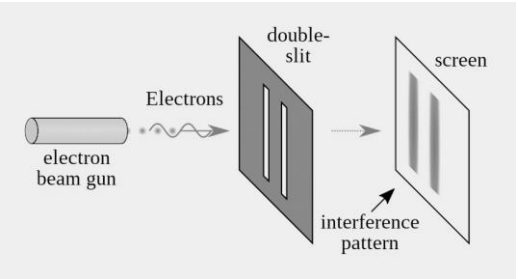

Figure 2: An illustration of an electron passing a double slit, not interfering with itself due to being observed. Image adapted from [3]

The conclusion of the double slit experiment is that the behaviors and characteristics of the electrons will appear differently depending on the circumstances like how we do our measurement [4]. This wave-particle duality makes the electron no longer best be described as either a particle or a wave but rather mix of both [5].

Superposition

These behaviors can be explained with the help of superposition. Meaning the electron can be in different states at the same time but not just as a statistical probability but that the different states can interfere with each other [4].

This can be described using Dirac notation. For a simple system in a superposition of two states the system can be expressed in the following way.

$$|\psi\rangle = a_1 \, |\psi_1\rangle + a_2 \, |\psi_2\rangle$$

Here $|\psi_1\rangle$ is one of the possible configurations of the state and $|\psi_2\rangle$ is the another.

An example to understand this superposition phenomena we can look at the interferometer (figure 3).

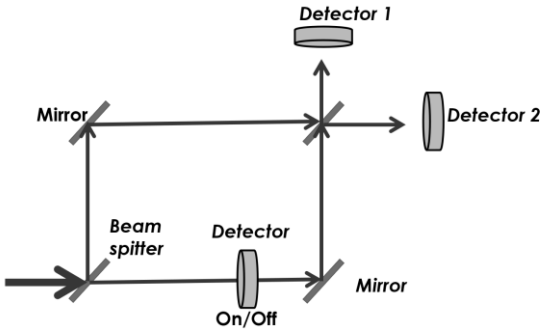

Figure 3: Illustration of an interferometer. Electrons coming from the left side and gets divided by the "beam splitter" getting in a super position. If the detector in the bottom is turned on, the electrons will not get into a superposition sins the detector will or will not detect the passage of the electron, making sure which one path the electron must have taken.

This interferometer consists of a beam that shoots electrons on to a beam splitter, sending the electrons into both directions in a superposition. The electron will then meet itself at the end and interfere with itself and thus be detected to detector 2 [6].

If we on the other hand would have a device, like a detector in one of the beam paths, that can tell us when an electron is passing by we will get a different scenario. The detector will in this case tell us that there was an electron passing it, or it will, by not seeing an electron passing, in a sense tell us that the electron must have taken the other path. This measurement makes the electron that was previously in a superposition, to be either in the path of the detector or the other and no longer in both simultaneously. This "collapse" of the superposition will be further explained in section about decoherence. But what this mean is two things. The measurement makes the electron lose its superposition and be in one of the two paths, and now being in one of the two paths it can no longer interfere with itself thereby will either get detected by detector 1 if the path was the one going up, or it will get detected by detector 2 if the path of the electron was the other [6].

Orbitals

The state of a single electron is usually referred to as an orbital.

In the region around a nucleus there are several types of orbitals with different symmetries. We will focus on two types of orbitals, the d-orbitals, and the p-orbitals.

In a cubic symmetry the d-orbitals can be combined into two different irreducible representations. These representations, or symmetrized orbitals, are called E_g and T_{2g}. There are two E_g orbitals and three T_{2g} orbitals, as illustrated in figure 4 [7].

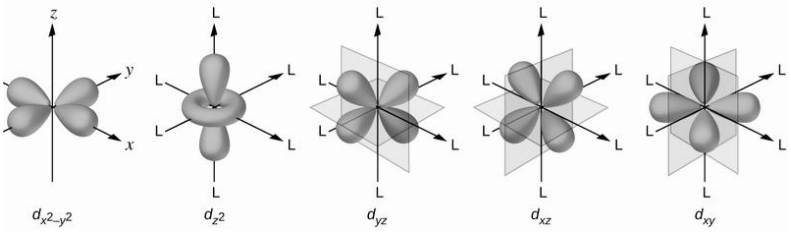

Figure 4: the 5 d orbitals. The two orbitals on the left have E_g symmetry and the three to the right have T_{2g} symmetry. Image adapted from [8]

Each orbital can be occupied by one electron with spin up and one electron with spin down, making the maximum number of electrons in the d orbitals being 10 electrons [7].

In a cubic symmetry, all three p orbitals belong to the same irreducible representation. Making the maximum number of electrons for the p-orbitals being six. If the cubical symmetry is broken for example by a magnetic field in the z direction, as it will be for this study, the orbitals can instead be divided into two other irreducible representation. They can be divided into two irreducible representations. P_x and p_y corresponds to the E_u symmetry and p_z corresponds to the A_u symmetry [9].

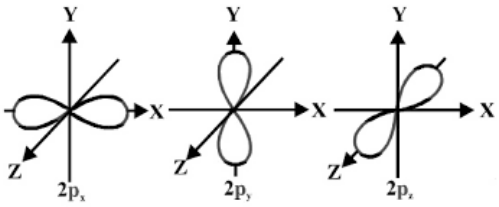

Figure 5: the 3 p orbitals. Image adapted from [10]

Many body states

When we have more than one electron, the electrons can occupy different orbitals and these different configurations corresponds to many-body states [11].

In classical physics we would be able to tell electrons apart from one another, but in quantum mechanics electrons are indistinguishable. This means that if two electrons where to switch places with each other, like in orbitals, this would not lead to a new many-body state but just change its sign.

To describe such systems, we introduce the creation and annihilation operators. $\hat{c}^+ , \hat{c}$. These operators can, just as the name suggest, be used to either add an electron to a state or remove from a state. We also introduce the Hamiltonian H_{ij} that governs the dynamics of the system through the Schrödinger equation [11].

The one particle Hamiltonian can be expressed as

$$\hat{\mathrm{H}} = \sum_{ij} H_{ij} \hat{c}_i^+ \hat{c}_j .$$

The Coulomb interaction between the two electrons can be expressed using U_{ijkl}

$$\hat{U} = \sum_{ijkl} U_{ijkl} \hat{c}_i^+ \hat{c}_j^+ \hat{c}_k \hat{c}_l .$$

In this term we have creation and annihilation operators for two electrons simultaneously and thus get the interaction between these.

Entanglement

For a many-body system we can divide the system into different subsystems. We can take the system of the d-orbitals and the p-orbitals for example and divide these systems into the d-orbital subsystem and the p-orbital subsystem. We could call the state of the d-electrons as $|\psi_d\rangle$ and the state of the p electrons as $|\psi_p\rangle$ [12] .

If these subsystems were to interact with each other or exist in near proximity to each other, however, we cannot describe these systems as independent systems, even if they later would to

be separated by a large distance. Their previous properties like superposition are now dependent on each other meaning a measurement or knowing the exact state of one system could lead to knowing the state of the other subsystem [13].

To take an example, let us consider the state the $|\psi_d\rangle$ to have two electrons, one with spin up and one with spin down in the same orbital $|½ - ½\rangle_d$ and the $|\psi_p\rangle$ to have no electron at the start, $|0\rangle_p$. Now we consider one of the electrons to exited and thus go from $|\psi_d\rangle$ to $|\psi_p\rangle$. Now we have a superposition of either a spin up in the d orbital and a spin down in the p-orbital of vise versa, as below [14]. (The a and b coefficients are the weights for the two superpositions).

$$|\psi_{dp}\rangle = a\,|½_d - ½_p\rangle + b\,|-½_d + ½_p\rangle$$

This state of the system can drastically change if we would be able to tell the spin one of the electrons for certain.

As many other quantum phenomena this entanglement is not something we experience in everyday life. To understand why we need to learn about measurements and Decoherence.

Decoherence

To understand the problem and reason for this study the key concept decoherence needs to be introduced and understood.

The Copenhagen interpretation vs Decoherence

One of the most common interpretations of QM is the Copenhagen interpretation. With it comes the perception that measuring or observing a quantum system makes it collapse, meaning that the measurement changes the wavefunction from a superposition to an eigenstate of the observed operator. This interpretation hence states that there is a state that describes the system before measurement and a state that describes the system after measurement linked by a special rule for the observer of a QM system.

A newer and more relevant interpretation of the observation of a QM system is the idea of decoherence and its effects on a QM system. To see the differences of these two we compare a scenario [12].

Let's take a QM system with an electron in an initial state described by a superposition of spin up and spin down $|\psi\rangle = a_1\,|\,½\,\rangle + a_2\,|-½\,\rangle$. In both interpretations this initial state would be

described in the same way but they will start to differ when we take an observer into account. In our Copenhagen interpretation the state of the QM system in a superposition would drastically change (collapse) under an observation of the spin. After the observation the state of the system is either $|\,½\,\rangle$ or $|\,-½\,\rangle$, depending on what is observed. The other term in the superposition is simply eliminated [16].

For the decoherence interpretation the perceived "collapse" is explained by a buildup of entanglement between the system and its environment. The buildup of entanglement can be seen as a leakage of information from the QM system to its environment. When the QM system interacts with its environment the state of the system and the state of the environment (env) gets for all practical purposes irreversibly entangled, as a realistic environment has an unlimited number of degrees of freedom. This could be written as $|\psi\rangle = a_1\,|\,½ + env_1\rangle + a_2\,|-½ + env_2\rangle$. The entanglement with the environment prevents the two states of the system to interfere with each other, if the corresponding states of the environment remain orthogonal. In other words, this explains the "collapse" as not an elimination of one of the possible states in the superposition, but instead that the states after the measurement are prevented from interfering with each other [12].

Closed and open systems

An isolated system is what we call a closed system. The isolation implies that the interaction between the system and its environment ($H_{system,env}$) is zero. In an open system this interaction is finite [12]. The environment, which we will from now on call the bath, has just like our system a Hamiltonian (H_{bath}). The Hamiltonian for the total system including the bath can be expressed as

$$H= H_{system} +H_{bath} + H_{system,bath}$$

For this study we have the scenario of a total system consisting of d-orbitals. p-orbitals and bath-orbitals. We have included Coulomb repulsion between the electrons in the d- and p-orbitals ($U_{d,\,d}$, $U_{p,\,p}$, and $U_{d,\,p}$), and a hybridization between the p-orbitals and the bath-orbitals ($H_{p,\,bath}$). The latter term allows the p-electrons to move from the p-orbitals to the bath-orbitals and back. The Hamiltonian for this total system can then be expressed as

$$H= H_d + H_{,p} +H_{bath} + U_{d,\,d} + U_{p,\,p} + U_{d,\,p} + H_{p,\,bath}$$

Screening

In our model we explicitly allow the electrons in one subsystem (the d-orbitals) to interact through the Coulomb interaction with the electrons in another subsystem (the p-orbitals). The effect of this interaction, on the state of one of the subsystems, is called screening. Screening gives two major effects.

The first effects is that when the electrons in the p-orbitals interact with the electrons in the d orbitals, the electrons in the d-orbitals appear effectively to interact much less. The reason is that due to the d-p-interaction a p-electron can leave the system when an additional electron enters the d-orbitals, so the total amount of electrons interacting with each other remain the same. If the d-p interaction is excluded the p-electrons would not compensate for the changes in the d-orbital occupation, thus leading to a different total repulsion [12]. One may effectively compensate for this lack of compensation by reducing (screening) the value of the $U_{d,d}$ term in the Hamiltonian.

The second effect of screening is that it can cause decoherence. The interaction between the p- and the d-electrons can make them entangled, and when the p-electrons leave the p-orbitals going into the bath they will still carry this entanglement. The d-electrons and the environment thus also become entangled which decoherence as previously described in section “The Copenhagen interpretation vs Decoherence”.

Current models that describe the electrons in a material do not explicitly take the entanglement between the screening and the screened electrons into account. However, by ignoring this effect one may overestimate the QM properties of the different subsystems [12].

Methodology

To determine how much decoherence our system suffers from we used a simulation of a full system consisting of a d-orbital system, a p-orbital system, and a bath. The code used for the simulations was called “ImpurityModel”. Alternations was done to the code to read and use bath state parameters for the p-orbitals, and additional code was written to calculate the linear entropy of the ground state which will tell us how much the quantum states are mixed. (see appendix “code for linear entropy”).

Pure states and Mixed states

If the system can be described with one state vector it is considered a pure state and pure states has zero entropy. For our study we look at the ground state of the total system, and since the total system is isolated the ground state is given by the (pure) eigenstate of the Hamiltonian with the lowest energy [2].

For the case of our study we will look at the electrons in the d-orbitals and see how entangled they are to the electrons in the environment (p-orbitals and bath). If we have entanglement between subsystems then it is not possible to express the state of one of the subsystems as a pure state, since the entanglement makes it depend on the other subsystem [2]. Instead, we need to describe the state of the subsystem with a density operator, as shown below.

The state of our full system expressed as

$$|\psi\rangle = \sum_{ij} \alpha_{ij} \ |\psi_i^d \psi_j^{P,Bath}\rangle$$

$$\sum_{ij} \left|\alpha_{ij}\right|^2 = 1$$

Let us consider the expectation value for an operator that only acts on the d-orbitals

$$\langle \hat{A}_{dd} \rangle = \langle \psi \ | \hat{A}_{dd} \ | \psi \rangle = \sum\nolimits_{ij\ i'j'} \langle \psi_i^d \psi_j^{P,Bath} \ | \ \alpha_{ij}^* \hat{A}_{dd} \alpha_{i'j'} \ | \psi_{i'}^d \psi_{j'}^{P,Bath} \rangle$$

Since the operator does not act on the environment ($\psi_j^{P,Bath}$) only the terms with identical p,bath states (j = j´) are non-zero, as different p,bath states are orthogonal.

$$\langle \hat{A}_{dd} \rangle = \sum_{ij\ i'} \langle \psi_i^d \psi_j^{P,Bath} \ | \ \alpha_{ij}^* \hat{A}_{dd} \alpha_{i'j'} \ | \psi_{i'}^d \psi_j^{P,Bath} \rangle$$

This can be rewritten using a partial trace

$$\langle \hat{A}_{dd} \rangle = Tr(\sum_{i'} \ |\psi_{i'}^d \psi_j^{P,Bath}\rangle \ \alpha_{i'j})(\sum_{i} \alpha_{ij}^* \ \langle \psi_i^d \psi_j^{P,Bath} \ |)\hat{A}_{dd})$$

$$= Tr(\sum_{i'} \ |\psi_{i'}^d\rangle \ \alpha_{i'j})(\sum_{i} \alpha_{ij}^* \ \langle \psi_i^d |\ )\hat{A}_{dd})$$

To easier see the next step we substitute the two sums with the states

$$\sum_{i'} |\psi_{i'}^{d}\rangle \alpha'_{i'j} | = |\tilde{\psi}^{j}\rangle \quad \text{and} \quad \sum_{i} \alpha'^{*}_{ij} \langle\psi_{i}^{d}| = \langle\tilde{\psi}^{j}|$$

We can normalize the states $|\tilde{\psi}^{j}\rangle$ with the following procedure

$$P_j = \langle\tilde{\psi}^{j}|\tilde{\psi}^{j}\rangle$$

$$|\psi^{j}\rangle = \frac{|\tilde{\psi}^{j}\rangle}{\sqrt{P_j}}$$

The probabilities P_j sums up to one

$$\sum_{j} P_j = \sum_{j} \langle\tilde{\psi}^{j}|\tilde{\psi}^{j}\rangle = \sum_{j} \sum_{i'} (\alpha_{ij}^{*} \langle\psi_{i}^{d}|) \left(\sum_{i'} \psi_{i'}^{d}\rangle \alpha_{i'j} \right)$$
$$= \sum_{j} \sum_{ii'} \delta_{ii'} \alpha_{ij}^{*} \alpha_{i'j} = \sum_{ij} |\alpha_{ij}|^{2} = 1$$

since $\langle\psi_{i}^{d}|\,\psi_{i'}^{d}\rangle = \delta_{ii'}$. Now we can define an expression for the density operator

$$\hat{\rho}_d = \sum_{j} P_j |\psi^{j}\rangle\langle\psi^{j}|$$

And thus, leave us with the expression

$$\langle\hat{A}_{dd}\rangle = Tr(\hat{\rho}_d \hat{A}_{dd})$$

Using the density operator $\hat{\rho}_d$ we can describe all expectations values for the d system. Since $\hat{\rho}_d$ mixes statistical probabilities (P_j) and quantum superpositions, the state it describes is called a mixed state. Telling if a state is mixed, and at what degree, the entropy of the state can be used.

Entropy

Now having found an expression for the density operator we are just one step from telling how much decoherence the system d is experiencing. This will be done by calculating how much entangled the d-system gets to the environment, in our case the Bath and the p system. This is done by tracing away the Bath and the p-orbitals from the total density matrix. If the subsystems are not entangled, then the states of the subsystems will remain pure. On the other hand, if the subsystems are entangled, the states of the subsystems will be mixed.

For a pure state the trace of its density matrix squared will be one, but for a mixed state, arising from partial trace of an entangled state, the trace of the density matrix squared will less than one. We can thus tell how much entanglement with the environment there is by calculating the linear entropy of the reduced system. For that we use the following equation for the linear entropy [2]

$$E[\hat{\rho}_d] = Tr(\hat{\rho}_d - \hat{\rho}_d^2)$$

The minimal model system

In our simulations the electrons in the p-orbitals are allowed to jump to the bath orbitals and back. The Bath orbitals consist of two parts, the valence bands which have a lower energy, and the conduction bands which have a higher energy. In addition, the valence bands and the conduction bands are also split into different irreducible representations A_u and E_u, just as the p-orbitals (see figure 6). Only the orbitals with the same irreducible representation hybridize.

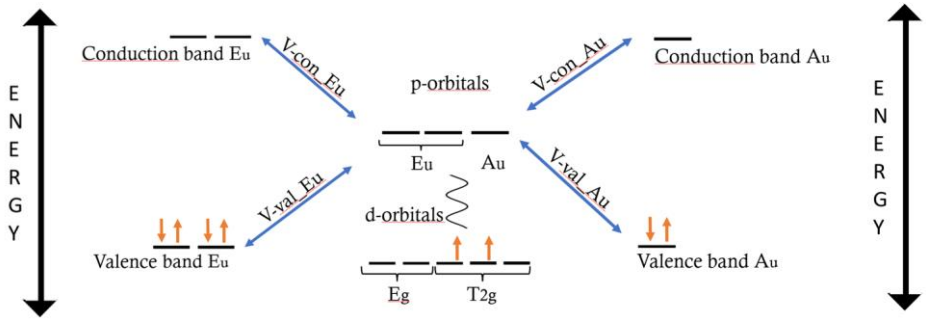

Figure 6: A visualization of the parameters and the orbitals of the Hamiltonian

To make sure the ground state is pure, the system is simulated at zero Kelvin and the parameters of the Hamiltonian are chosen in such a way that the eigenstate of the Hamiltonian with the lowest energy is non-degenerate. Degenerated states are when more than one state has the same energy. If that is the case, we can no longer tell states apart by looking at the energy levels and there by risking measuring a bigger effect of decoherence than what there actually is.

Hamiltonian of the model system

To make this simulation a Hamiltonian of the system has to be constructed. Previous in section "closed and open systems" we considered the Hamiltonian as such,

$$H = H_d + H_p + H_{bath} + H_{p,bath} + U_{dd} + U_{pp} + U_{dp}$$

For the sake of presenting these terms in a structured way, we group them in the following way.

$H_0 = H_d + H_p + H_{bath} + H_{p,bath}$ And $U_0 = U_{dd} + U_{pp}$

In order to lift the degeneracy of the ground state, we introduce two additional terms, a week spin-orbit coupling term $\hat{\mathrm{H}}_{\mathrm{so}}$ for the d-orbitals (ξ = 0.01 eV) and a magnetic term $\hat{\mathrm{H}}_{\mathrm{B}} = B \cdot \hat{S}_z$ ($B = 0.05$). Thus the final and total Hamiltonian is

$$\hat{H} = H_0 + U_0 + \hat{H}_{so} + \hat{H}_B$$

Now we will take a deeper look into all the terms of the Hamiltonian.

Parameters for the data, H_0 Hamiltonian and their meaning

Figure 6 gives an overview of the different orbitals in the system. Figure 7 shows the matrix representation of the one-particle Hamiltonian H_0. The E_u and the E_g representations both contain two orbitals each, while the T_{2g} contains three orbitals. The full definition for the parameters are defined in the table below.

$$H_0 = \begin{pmatrix} \varepsilon_{Au} & 0 & 0 & 0 & v_{Au}^{val} & 0 & v_{Au}^{con} & 0 \\ 0 & \varepsilon_{Eu} & 0 & 0 & 0 & v_{Eu}^{val} & 0 & v_{Eu}^{con} \\ 0 & 0 & \varepsilon_{Eg} & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & \varepsilon_{T2g} & 0 & 0 & 0 & 0 \\ \multicolumn{2}{c}{\multirow{2}{*}{Hc}} & 0 & 0 & \varepsilon_{Au}^{val} & 0 & 0 & 0 \\ & & 0 & 0 & 0 & \varepsilon_{Eu}^{val} & 0 & 0 \\ \multicolumn{2}{c}{\multirow{2}{*}{Hc}} & 0 & 0 & 0 & 0 & \varepsilon_{Au}^{con} & 0 \\ & & 0 & 0 & 0 & 0 & 0 & \varepsilon_{Eu}^{con} \end{pmatrix}$$

p-orbitals

d-orbitals

bath orbitals

Figure 7: the Hamiltonian as the impurity model reads it in matrix form

Name in dataset	Name in H^0	Description
A_u	ε_{Au}	The energy level for the p orbital with symmetry A_u
E_u	ε_{Eu}	The energy level for the p orbitals with symmetry E_u
E_g	ε_{Eg}	The energy level for the d-orbitals with symmetry E_g
T_{2g}	ε_{T2g}	The energy level for the d-orbitals with symmetry T_{2g}
Valence band E_u or e-val E_u	ε_{Eu}^{val}	The energy level for the valence band with the **E_u** symmetry
Conduction band E_u or e-con E_u	ε_{Eu}^{con}	The energy level for the conduction band with the **E_u** symmetry
Valence band A_u or e-val A_u	ε_{Au}^{val}	The energy level for the valence band with the **A_u** symmetry
Conduction band A_u or e-con A_u	ε_{Au}^{con}	The energy level for the conduction band with the **A_u** symmetry
v-con_ E_u	v_{Eu}^{con}	Strength of the hopping from the conduction band to the p-orbitals with **E_u** symmetry
v-val_ E_u	v_{Eu}^{val}	Strength of the hopping from the valence band to the p-orbitals with **E_u** symmetry
v-con_ A_u	v_{Au}^{con}	Strength of the hopping from the conduction band to the p-orbital with **A_u** symmetry
v-val_ A_u	v_{Au}^{val}	Strength of the hopping from the valence band to the p-orbital with **A_u** symmetry

The $\hat{U}$ interaction parameter

This is the coulomb interaction parameter that allows the electrons to interact with each other.

$$\hat{U} = \sum_{ijkl} U_{ijkl} \hat{c}_i^+ \hat{c}_j^+ \hat{c}_k \hat{c}_l$$

U_{dd}: Coulomb interaction between the electrons in the d-orbitals.

U_{pp}: Coulomb interaction between the electrons in the p-orbitals.

U_{dp}: Coulomb interaction between the electrons in the p-orbitals.

Selection of parameters

In materials with partially filled 3d orbitals the occupation of the 4p-orbitals is realistically somewhere between 0.3 and 0.7. The parameters of the model were selected to keep the p-orbital occupation close to this range.

It is also common that there is a slight difference in the energy levels for the T_{2g} and E_g orbitals. In our model we set $\varepsilon_{E_g} - \varepsilon_{T_{2g}} = 0.5$ eV so that the T_{2g} orbitals have a lower energy compared to the E_g.

Code for linear entropy

The function that was additionally written to the "impurity model" code was named "linear entropy" and can be found in the appendix with comments. This function calculates the entropy for the d-orbital system by first constructing thee reduced density matrix and then apply the formula for the linear entropy.

Impurity model Code

The "impurity model" is used to calculate the ground states. The main code can be fund with this GitHub link (https://github.com/JohanSchott/impurityModel)

Note: The code fund by the "GitHub link" is an unaltered edition and will not include the "linear entropy" function nor the minor alterations to the "impurity model" code.

Results

Note that there are a lot of parameters that effect the data and therefor some are selected to be fixed and others to vary to get some grasp of what causes high respectively low decoherence. The decoherence is as previous stated measured using the entropy for the reduced density matrix $\hat{\rho}_d$. Zero entropy means that the ground state is a superposition of infinitely many states, each with maximal entanglement between the subsystem.

Decoherence induced by the A_u bath orbitals

Let's first consider a minimal system composed of the d- and p-orbitals, and two bath orbitals with A_u symmetry. The bath orbtials with E_u symmetry are detached from the system ($v_{Eu}^{val} = v_{Eu}^{con} = 0$). The two A_u bath orbitals are separated in energy to represent a valence band and a conduction band., as shown in figure 6.

Table 1

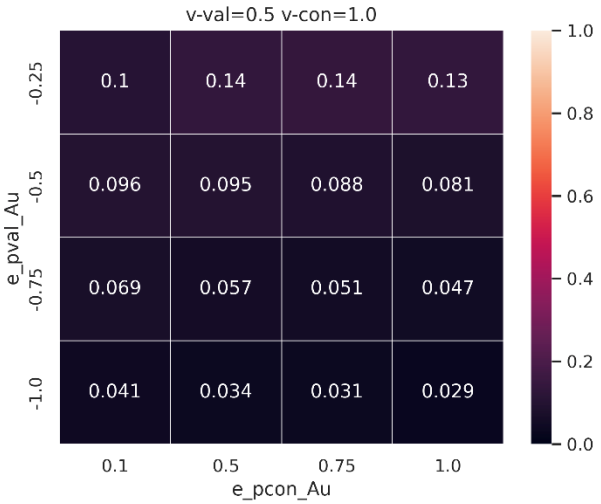

Figure 8: Measured entropy for selected parameters.

Table 1 shows the entropy of $\hat{\rho}_d$ when the hopping to the valence band is set to 0.5 and the hopping for the conduction band is set to 1.0. With higher conduction band the entropy decreases with one exception for when the valence energy is -0.25 eV. An explanation for this overall trend could be that the hopping from the p-orbital to the conduction band is suppressed when the conduction band energy is too high. This would effectively reduce the number of degrees of freedom in the environment. For the valence band we can see that with lower energy the entropy decreases. A reason for this might be that the hopping from the valence band to the p-orbitals is suppressed when the valence band energy is lowered. Overall, the decoherence is not very high. An explanation for this could be that the p-electrons with A_u symmetry are not able to distinguish the different d-electron configurations.

Table 2

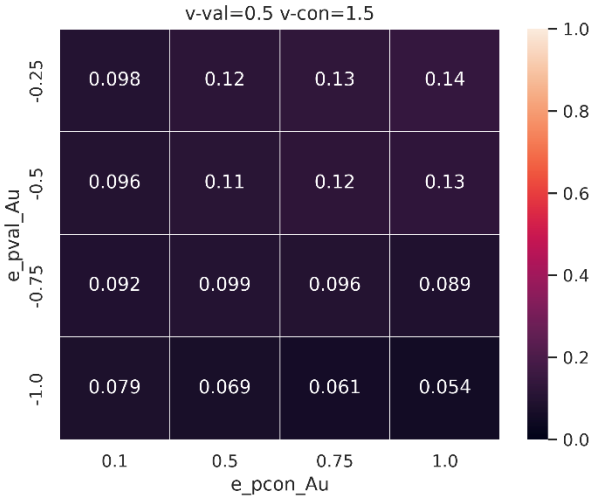

Figure 9: Measured entropy for selected parameters

Table 2 shows the entropy for system with $v_{Au}^{val} = 0.5$ eV $v_{Au}^{con} = 1.5$ eV. Similar to the case with $v_{Au}^{con} = 1.0$ eV, when the valence band energy is at a lowered the decoherence decrease. An explanation of this would be that the hopping from the valence band is suppressed when the energy level is low, resulting in the p-electrons not moving around that much and thus not spreading the information to the environment.

The most interesting thing for this set of systems with hopping strength $v_{Au}^{con} = 1.5$ eV (Table 2) compared to t $v_{Au}^{con} = 1.0$ eV (Table 1) is that for valence band energy between -0.25 eV and -0.5 eV the decoherence increases when the energy level for the conduction band is raised . The opposite is observed when the valence band is between -0.75 eV and -1.0 eV. Here we see, much like in table 1, a decrease in decoherence when the conduction energy is raised. For the former trend one can speculate that the conduction band and the p-orbital form a bonding orbital (a superposition between the p-orbital and the bath orbitals that has low energy in H_0) with such low energy that it is almost fully occupied in the ground state.

Decoherence induced by the E_u bath orbitals

Now we consider a minimal system composed of the d- and p-orbitals, and four bath orbitals with E_u symmetry. This time the bath orbitals with A_u symmetry are detached from the system ($v_{Au}^{val} = v_{Au}^{con} = 0$). The four E_u bath orbitals are separated in energy to represent a valence band and a conduction band, as shown to the left in figure 6.

Table 3

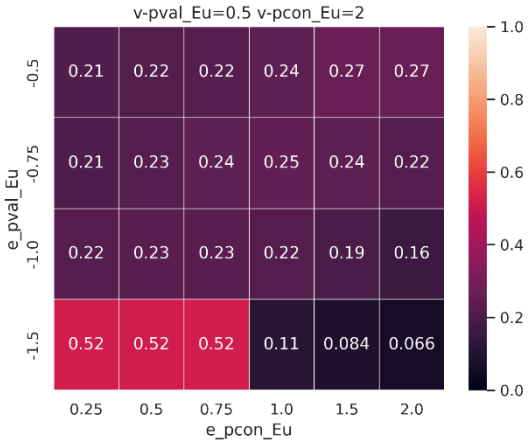

Figure 10: Measured entropy for selected parameters

Table 3 shows the entropy for this system when the energies for the valence and conduction bath-orbitals are varied.

Comparing this table with table 1 and 2 it is noteworthy that there is an overall stronger decoherence effect. This can be explained by the increase in the degrees of freedom of the environment (p-orbitals and bath) since there are two orbitals in the E_u irreducible representation and only one in the A_u.

In this example we have a very high decoherence when we gave a low valence level and conduction level. We can see a very large decoherence when ε_{Eu}^{val} = -1.5 eV and ε_{Eu}^{con} = 0.25 to 0.75 eV. By analyzing the ground state for these cases, we see that they are clearly different compared to the ground state of the other cases mainly by the large variation of different d- and p-configurations.

Decoherence induced by both the A_u and E_u bath orbitals

Finally, we look at the combined decoherence effect of having bath orbitals with both A_u and E_u symmetries. The fixed parameters for this setup are $v_{Au}^{val} = 0.5$, $v_{Eu}^{val} = 0.5$, $v_{Au}^{con} = 1.0$, $v_{Eu}^{con} = 1.5$.

Table 4

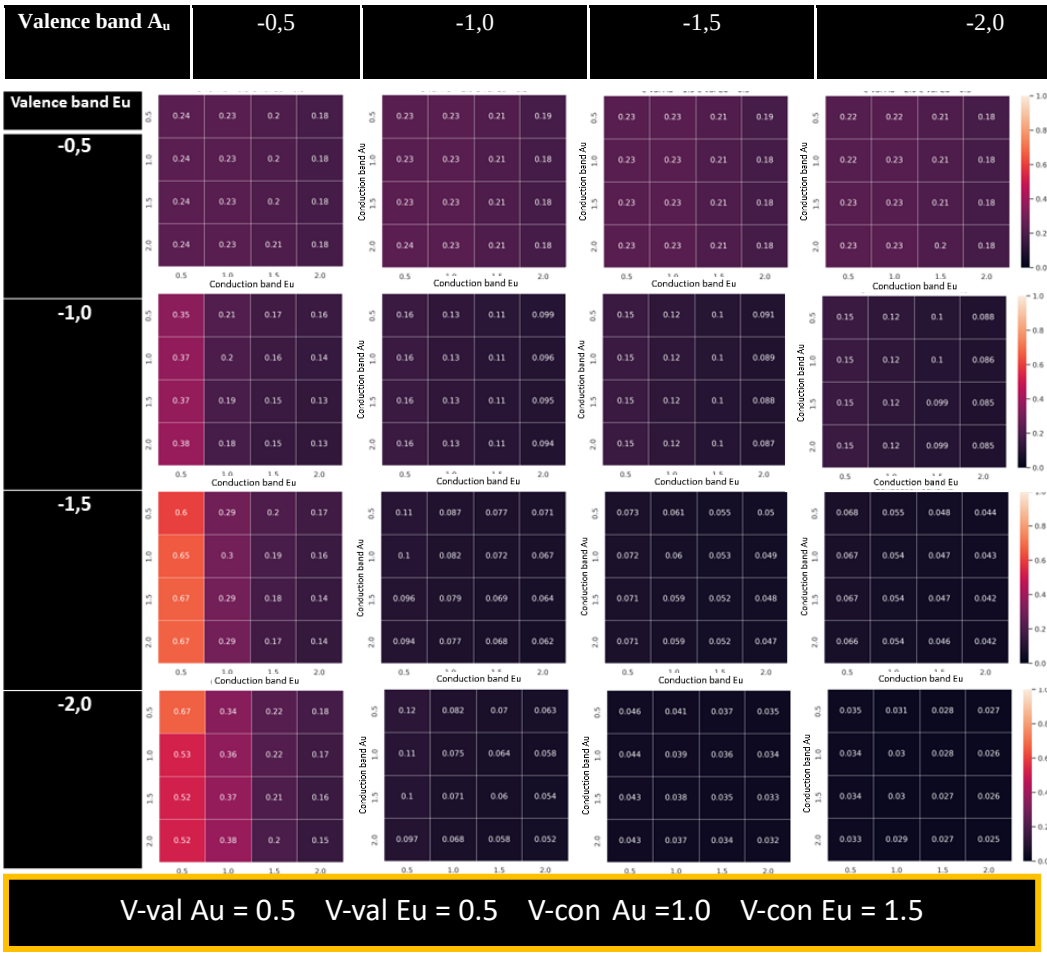

Figure 11: Measured entropy for selected parameters. Image from [7]

Table 4 consist of 16 plots of the entropy for systems with different values of the bath orbital energies.

In the considered energy range, when $\varepsilon_{Au}^{val} \leq \varepsilon_{Eu}^{val}$ the entropy is almost independent of ε_{Au}^{val}. In this region the decoherence decrease as ε_{Eu}^{val} is lowered. It is noteworthy that the occupation of the p-orbital remains around 0.5 electrons throughout this region.

For the region where $\varepsilon_{Au}^{val} \geq \varepsilon_{Eu}^{val}$ the situation changes drastically. Here we see a strong increase in decoherence when ε_{Au}^{val} = -0.5, ε_{Eu}^{val}=-1.5, and ε_{Eu}^{con}= 0.5. These parameters are similar to those that gave a high entropy in table 3. However, in table 4 we see that the inclusion of the A_u bath orbitals strongly effects the entropy. When the valence band for A_u is lowered the entropy changes abruptly from 0.67 to 0.1. This is due to a large change in the ground state.

One physical effect that could play a role is the repulsion between the electrons in the different p-orbitals with different irreducible representation. This repulsion affects the decoherence in a

way that is not only additive since it will lower the contribution from the states with more than one p-electron.

The dependence of electrons in p-orbitals

One idea for explaining the amount of decoherence was that it could depend on the number of p-electrons. A quick study of this was made to see if there are any clear trends for the number of p-electrons and the amount of decoherence of the system. In figure 12, 13, 14, and 15 the entropy is plotted against the amount of electrons in the p-orbitals for various cuts through the parameter space.

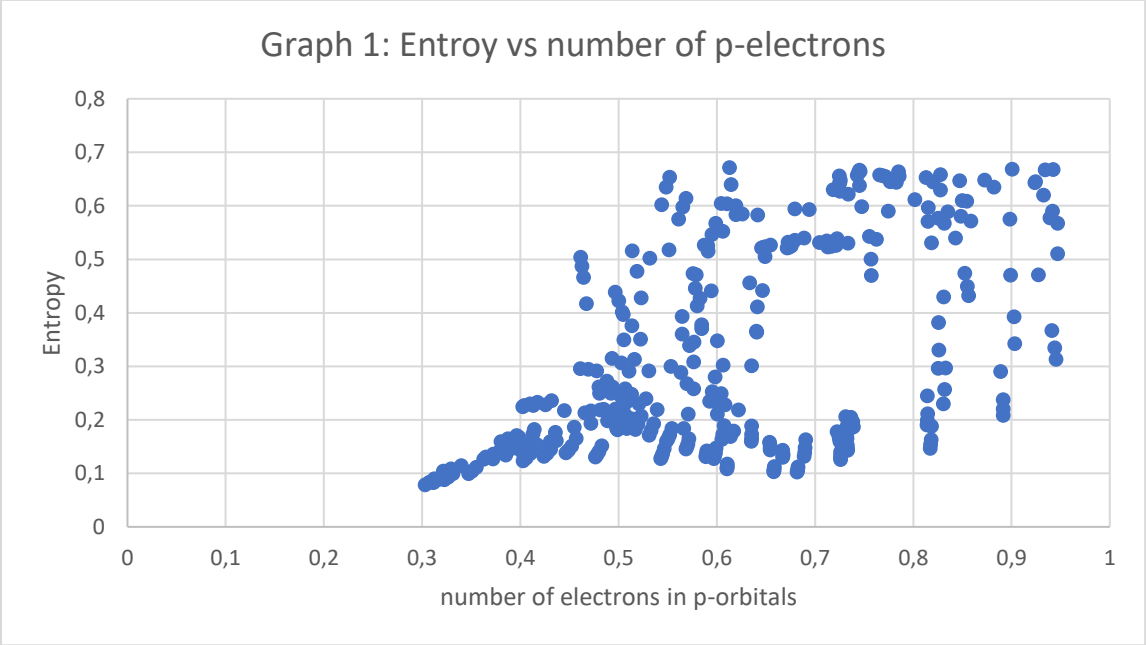

Figure 12: Entropy plotted against number of p-electrons for the parameter cut in eV: v_{Au}^{val} = 0.5, v_{Eu}^{val}= 0.5, v_{Au}^{con}= 1 to 2, v_{Eu}^{con}= 1 to 2, ε_{Au}^{val}=-0.25 to -0.75, $\varepsilon_{Eu}^{con} = -1.5$ ε_{Au}^{con}= 0.1 to 1, ε_{Eu}^{con}= 0.1 to 1.

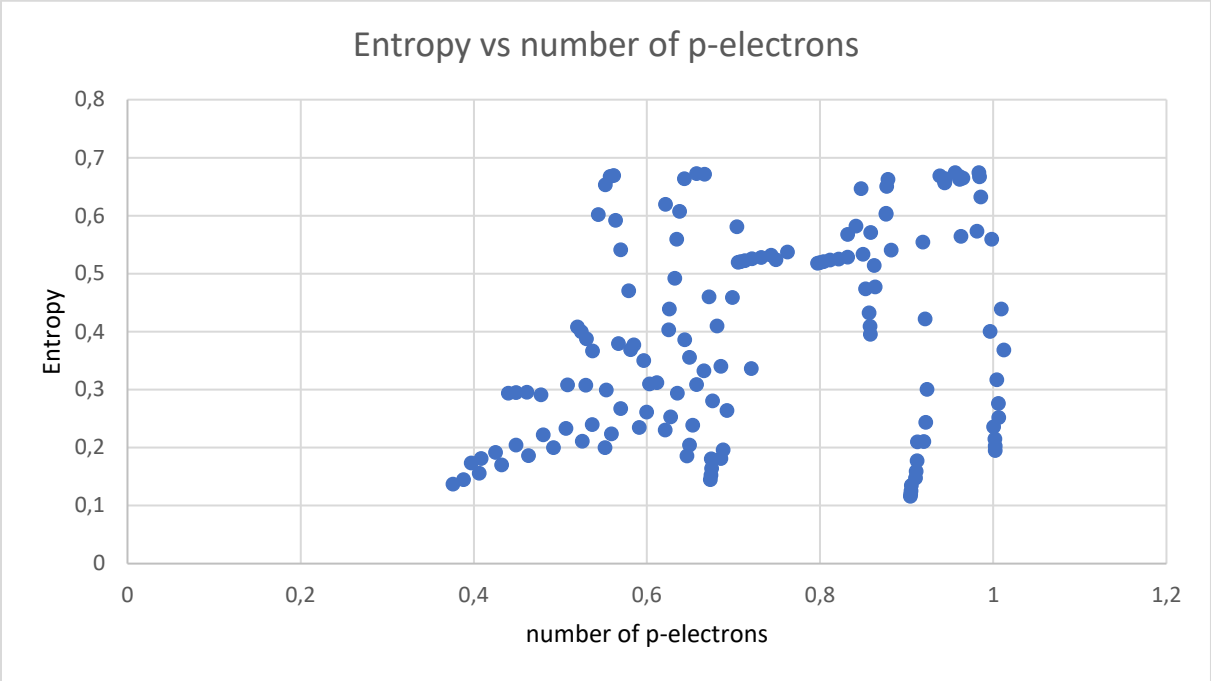

Figure 13: Entropy plotted against number of p-electrons for the parameter cut in eV: v_{Au}^{val} = 0.5, v_{Eu}^{val}= 0.5, v_{Au}^{con}= 1 to 2, v_{Eu}^{con}= 1.5 to 2.5, ε_{Au}^{val}=-0.5, $\varepsilon_{Eu}^{con} = -1.5$, ε_{Au}^{con}= 0.5 to 2, ε_{Eu}^{con}= 0.5 to 2.

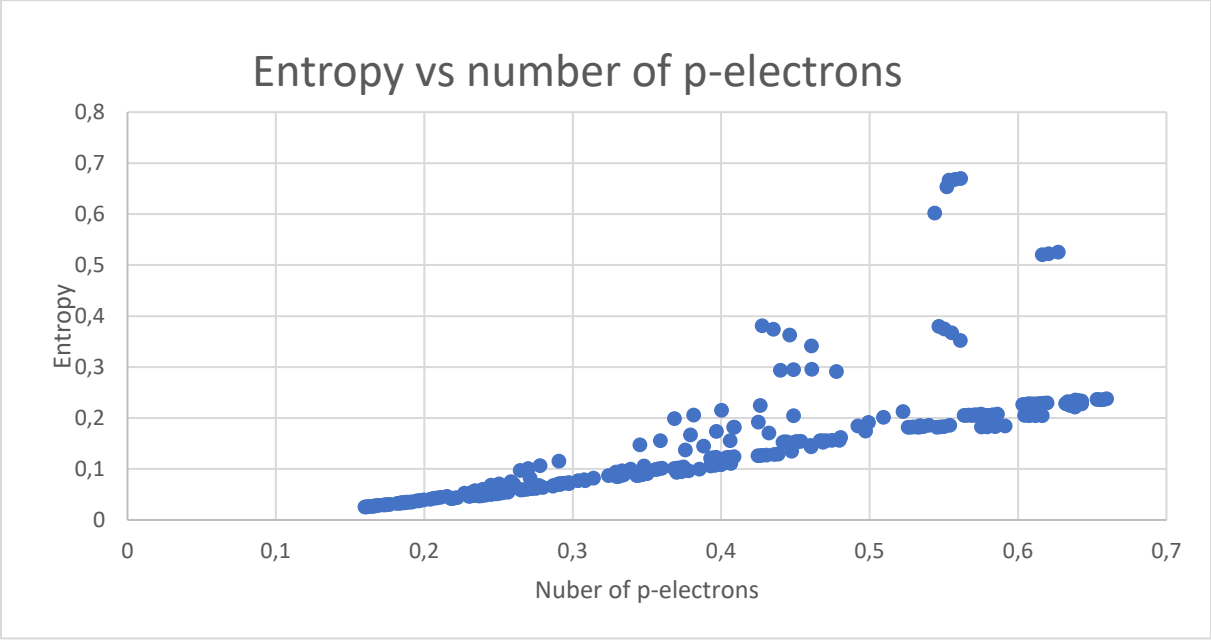

Figure 14: Entropy plotted against number of p-electrons for the parameter cut in eV: v_{Au}^{val} = 0.5, v_{Eu}^{val}= 0.5, v_{Au}^{con}= 1, v_{Eu}^{con}= 1.5 to 2, ε_{Au}^{val}=-0.5 to -2, $\varepsilon_{Eu}^{con} = -0.5$ to -2, ε_{Au}^{con}= 0.5 to 2, ε_{Eu}^{con}= 0.5 to 2.

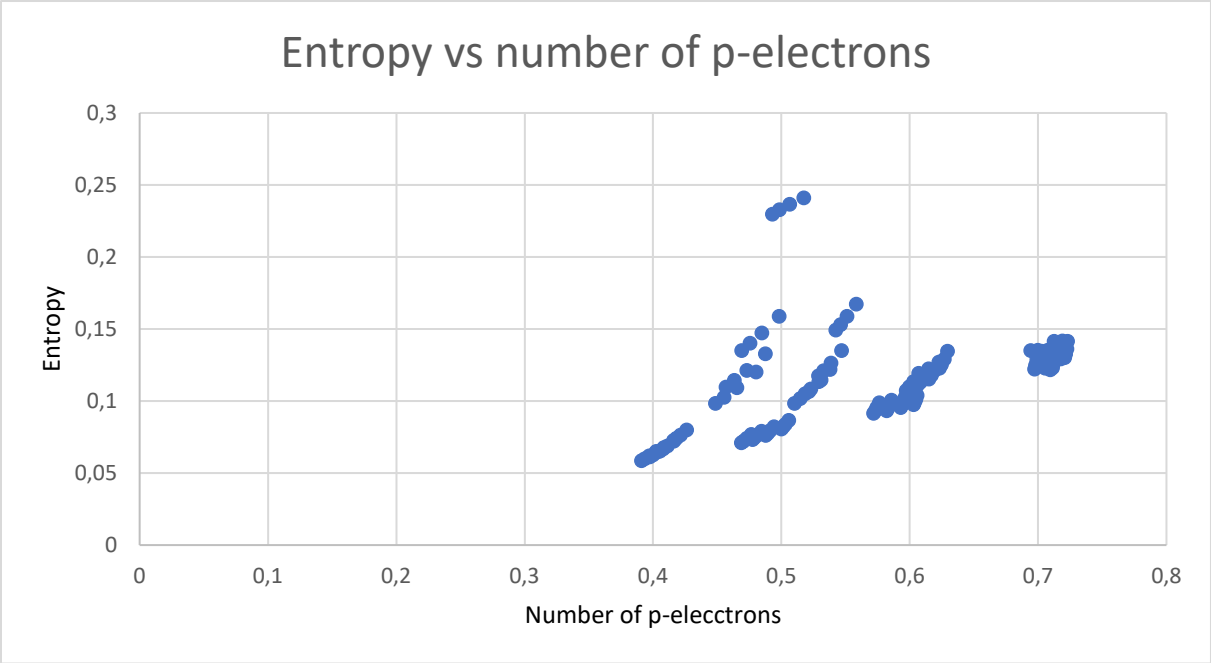

Figure 15: Entropy plotted against number of p-electrons for the parameter cut in eV: v_{Au}^{val} = 0.5, v_{Eu}^{val}=1, v_{Au}^{con}= 1, v_{Eu}^{con}= 1.5, ε_{Au}^{val}=-0.5 to -1.5, $\varepsilon_{Eu}^{con} = -0.5$ to -2 ε_{Au}^{con}= 0.5 to 2, ε_{Eu}^{con}= 0.5 to 2.

In all cuts the amount of decoherence (measured entropy) is the highest when the number of electrons occupying the p-orbitals is 0.5 and higher but having a high number of p-electrons does not necessarily imply a high amount of decoherence, as seen in figure 14 and 15. For an p-orbital occupation lower than 0.4 there seems to be a clear linear trend between entropy and occupation number. Figure 12 and 13 seems to indicate that when the number of p-electrons exceeds 0.55 the decoherence won't go higher but instead finds some sort of limit close to 2/3. The eigenvalues of the reduced density matrix shows that this upper limit is due to that the d-electrons in the ground state primarily take three different configurations, which implies $Tr(\hat{\rho}_d^2) \leq 1/3$.

Discussion

That there would be decoherence of some magnitude was expected in a way but the high amount of it is very surprising. The system simulated is of a minimal model with a limited description of the environment, but still with properties similar to that of a transition metal atom in a realistic material. Even with this limited number of degrees of freedom the bath was still able to resolve the different configurations of the d-electrons to a large extent.

The aim of the study was to quantify the amount of decoherence induced by the p-d-electron interaction to see if it can be neglectable in realistic models of materials. The result of this study shows that for some parameter regions it is not a good approximation to neglect the decoherence effect. What has been found is that p-d-electron interaction surely is something that needs to be investigated further to make more accurate approximations.
From the data it seems like the bath orbitals with E_u symmetry are important to induce a large amount of decoherence. When this symmetry is active there is a higher number of degrees of freedom in the environment, which is needed to resolve the different d-configurations.

An interesting direction for future studies is trying to understand the physical mechanisms that amplify the decoherence in order tell when it can be neglected or not. For example, a physical mechanism explored was how the number of p-electrons relate to the entropy of the reduced density matrix. The figure 12-15 says that the number of p-electrons is not what necessarily gives of a high amount of decoherence.

To expand on this study, one could expand the model system with bath orbitals that hybridize directly with the d-orbitals. Another addition could be to consider f-orbitals instead of d-orbitals and exchanging the p-orbitals with d-orbitals. Varying the number of electrons in the different subsystems. Extending the model system in this way would make the calculations much heavier. The runtime for the simulation would go from minutes to days and thus become unsuitable for a bachelor thesis.

Conclusion

The main conclusion of this study is that the decoherence effect from the interaction between the p- and d-electrons cannot be neglected. This highlights the need for continued method development to take this effect into account in realistic material simulations.

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Appendix

Code for linear entropy

```
def linear_entropy(nBaths,n_spin_orbitals,psi):
#Finds  egenvalues for the Hamiltonan (ws) and calculate the absolute walue. putting these in a new
# list as a tupple
    ws = np.array([ a for a in psi.values() ])
    absws = np.array([ abs(a)**2 for a in psi.values() ])
    s = np.array([ psr.bytes2tuple(ps,n_spin_orbitals) for ps in psi.keys() ])
    j = np.argsort(absws)
    ws = ws[j[-1::-1]]
    s = s[j[-1::-1]]
#constructing two list. one for p-states+Bath (PB)states and one for the d-states(D). This is to later
#find the superpossitions fo d-states. This will constists of lists with lists as elements.
    nps = 0
    PB=[]
    D=[]
    for i,slate in enumerate(s):
        npelec = 0
        d=[]
        pb=[]
        for c in slate:
            if 5<c<16:
                d.append(c-6)
            else:
                pb.append(c)
                if c < 7:
                    npelec = npelec + 1
        nps = nps + npelec*np.abs(ws[i])**2
        D.append(d)
        PB.append(pb)
#Sorting the p Bath list in order and the same order for the weigt for the states and the d list.
    isort = sorted(range(len(PB)), key=PB.__getitem__)
    PB = [PB[i] for i in isort]
    D = [D[i] for i in isort]
    ws=[ws[i] for i in isort]
    wslist = []
#finding the superpossitions for d where the p bath is identical.
```

```
    D_Trace=[]
    ws_Trace=[]
    for i in range(len(PB)-1):
        dlist.append(D[i])
        wslist.append(ws[i])
        if PB[i] != PB[i+1]:
            D_Trace.append(dlist)
            ws_Trace.append(wslist)
            dlist = []
            wslist = []
    dlist.append(D[-1])

    wslist.append(ws[-1])
    D_Trace.append(dlist)
    ws_Trace.append(wslist)
#Calculation of the dencity matrix
    n_electrons = len(D_Trace[0][0])
    n_space = 10
    rho = np.zeros((n_space**n_electrons, n_space**n_electrons),dtype=np.complex)
    rhovec = np.zeros((n_space**n_electrons),dtype=np.complex)
    numvec = [n_space**(n_electrons-k-1) for k in range(n_electrons)]
    for i, state in enumerate(D_Trace):
        rhovec[:] = 0
        for j, slater in enumerate(state):
            b = np.dot(slater, numvec)
            rhovec[b] = ws_Trace[i][j]
    rho += np.outer(rhovec,np.conjugate(rhovec))


#Calculating the linear entropy


    print("Tr(rho) = ",np.trace(rho))
    print("Tr(rho^2) = ",np.trace(rho.dot(rho)))
    entropy = np.trace(rho - rho.dot(rho))
    return entropy,nps
```