Simulation of ultra fast magnetization dynamics

Document status and date:
Published: 01/01/2008

Document Version:
Publisher's PDF, also known as Version of Record (includes final page, issue and volume numbers)

Please check the document version of this publication:

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Simulation of ultra fast magnetization dynamics

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December 6, 2008

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Chapter 1

Introduction

In the late nineteenth century the frenchman Pierre Curie discovered the fact that the magnetic field exhibited by a ferromagnetic material decreases when its temperature is raised, and recovers its magnetic field when cooled again. This phenomena is of great importance in modern day computer harddisk technology. Because of continual demand for higher capacities and speed it is necessary that the small magnetic elements in a harddisk (that represent the information) can be switched faster and more efficiently. This can be achieved by heating the magnetic element, such that the field needed to switch the element is much lower than before. This allows for much faster switching and reduces the chance that nearby elements are affected as well. Because of the small size of the elements, the heating must be very local and precise. In order to accomplish this feat, a laser can be used. This laser induced demagnetization will be the main focus of this report.

The macroscopic experiment that lies at the basis is schematically depicted by:
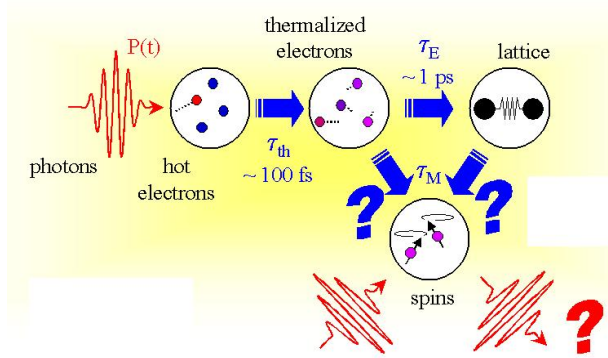
Figure 1.1: Laser induced demagnetization.

A short laser pulse hits the material and excites the electrons to higher energy states. After a certain time these hot electrons have reached an internal equilibrium again. This thermalization process happens relatively fast. Now these thermalized electrons are no longer in equilibrium with the lattice. As a result they will start to interact and achieve a new equilibrium state. This however occurs with a much higher timescale τ_e of about $1ps$. During this time the magnetization of the sample, which can be measured by using a MOKE-setup (Magneto-Optical Kerr Effect), will also change. The timescale τ_m that belongs to this demagnetization process is generally believed to be higher than τ_e. What we intend to show is that it is

possible for the demagnetization to go faster than the electron-lattice equilibration. Furthermore we will investigate the possibility of magnetic switching by applying an external field. In the next chapter we will present a model that is capable of simulating this experiment.

Chapter 2

The model

To be able to model the experiment described in the previous chapter it is first necessary to understand exactly what systems and interactions are involved. They will be explained separately in the next sections. After that we will look at some basic simulations in order to check if the model we derived is indeed valid. It is important to realize that it will be a deterministic model that is entirely based upon looking at what happens to "the average atom".

2.1 Subsystems

2.1.1 Electrons

The electrons are treated as a Fermi sea of spin-less particles. We define this subsystem by its quantum mechanical distribution of electronic states. In terms of the model this means we take a certain energy range around the Fermi-level and divide it into a number of cells (see figure 2.1 below). For a detailed mathematical analysis of such a discrete system of electrons we refer the reader to appendix B.
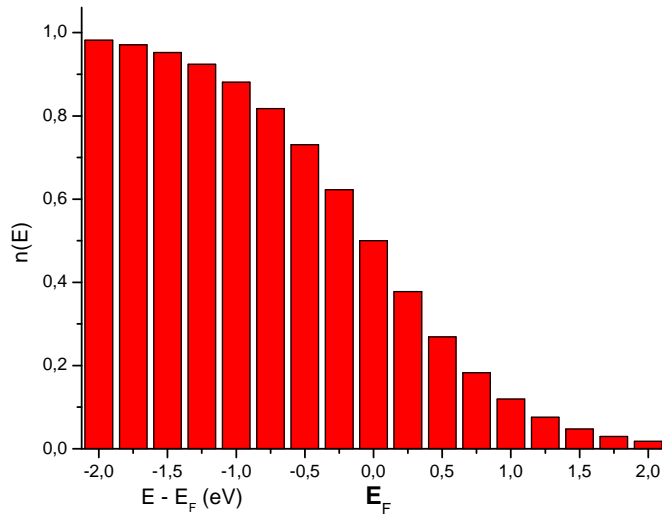

Figure 2.1: Representation of the electronic system.

Each cell with energy E_i is assigned a number n_i between 0 and 1 representing the relative occupation of states which are within the energy range of that cell. The total number of electrons N_i in a certain cell is then given by:

$$N_i = n_i D_e \Delta E \tag{2.1}$$

where D_e is the density of states which is assumed constant for all energy levels, and ΔE the energy width of a cell. The total energy E_e (per atom) of the electrons is:

$$E_e = \sum N_i E_i \tag{2.2}$$

In equilibrium at temperature T_e the occupation of these electronic states is described by the Fermi-Dirac distribution:

$$n_i = \frac{1}{e^{\frac{E_i}{k_B T_e}} + 1} \tag{2.3}$$

The Fermi level is taken to be 0 so E_i can be negative. From equation 2.3 we can derive an expression for the electron temperature T_e. If we take the derivative with respect to E_i we get:

$$\frac{\partial n_i}{\partial E_i} = -\frac{1}{(e^{\frac{E_i}{k_B T_e}} + 1)^2} \frac{1}{k_B T_e} e^{\frac{E_i}{k_B T_e}} \tag{2.4}$$

which means $\left. \frac{\partial n_i}{\partial E_i} \right|_0 = \frac{1}{4 k_B T_e}$. This gives us:

$$T_e \equiv \frac{1}{4 k_B \left. \frac{\partial n_i}{\partial E_i} \right|_0} \tag{2.5}$$

Of course this temperature only has a meaning when the electron system is in equilibrium. In a non-equilibrium situation we have to specify all the n_i to completely describe the state of the system.

A last and useful parameter is the electronic heat capacity c_e which is defined as the derivative of the total electron energy E_e with respect to the temperature T_e. In the continuous case an expression for c_e can be derived but we will not show it here. The result is:

$$c_e \equiv \frac{\partial E_e}{\partial T_e} = \frac{1}{3} \pi^2 k_B T_e \tag{2.6}$$

2.1.2 Lattice

The lattice consists of a system of coupled harmonic oscillators, obeying Bose-Einstein statistics. We assume a number of D_p oscillators per atom with an energy spacing (also called phonon energy) of E_p (see figure 2.2 below).
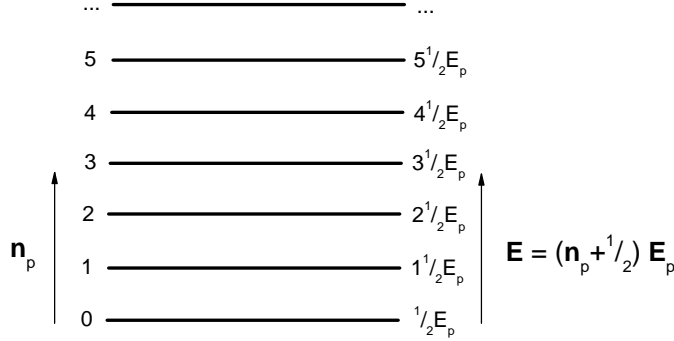
Figure 2.2: Energy levels of an harmonic oscillator.

In equilibrium the average number of phonons n_p per oscillator is given by:

$$n_p = \frac{1}{e^{\frac{E_p}{k_B T_l}} - 1} \tag{2.7}$$

where T_l is the temperature of the lattice.

The total number of phonons per atom N_p is then:

$$N_p = n_p D_p \tag{2.8}$$

Equation 2.7 relates n_p to T_l so this means only one of these two is necessary to define the state of the lattice. We usually take the one that is easiest to use in a certain situation. Another important parameter is the heat capacity c_l (per atom). It relates the temperature T_l to the total energy (per atom) E_l that is stored in the lattice:

$$E_l = c_l T_l \tag{2.9}$$

For simplicity we assume c_l is constant.

A last thing to mention is that the lattice is considered to continuously remain in equilibrium, i.e. we assume infinitely fast phonon-phonon interactions.

2.1.3 Spin-system

The spin of the electrons is treated in the simplest way possible where an electron can be either in the spin-up $(+\frac{1}{2})$ or spin-down state $(-\frac{1}{2})$. All spins are chosen to be aligned in the same direction. We assume an number of D_m effective spins per atom, and we keep track of the average spin s. We call the energy difference between the spin-up and spin-down state E_m (see figure 2.3).

We use the Weiss model for magnetism so E_m is considered to be dependent on s:

$$E_m(s) \equiv E_{\sigma\uparrow} - E_{\sigma\downarrow} = -2sE_{m0} \tag{2.10}$$
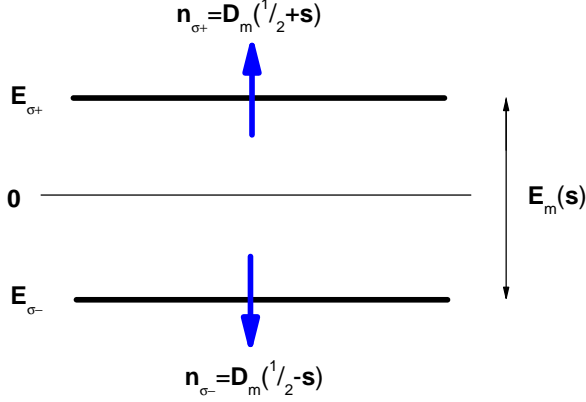
Figure 2.3: Representation of the spin-system.

where E_{m0} is defined as the difference in energy at $s = -\frac{1}{2}$. We would also like to introduce a temperature definition. To this end we assume that the spin-system follows Boltzmann statistics in the following way.

The number of spins in a particular state can be calculated from the average spin s as follows:

$$n_{\sigma\uparrow} = D_m(\frac{1}{2} + s)$$

$$n_{\sigma\downarrow} = D_m(\frac{1}{2} - s)$$

In equilibrium these two numbers will be related by:

$$\frac{n_{\sigma\uparrow}}{n_{\sigma\downarrow}} = e^{-\frac{E_m}{k_B T_m}} \tag{2.11}$$

where T_m is the spin temperature. If we rewrite the above expression we get:

$$T_m \equiv -\frac{E_m}{k_B \ln \frac{\frac{1}{2}+s}{\frac{1}{2}-s}} \tag{2.12}$$

At $T_m = 0$ we take $s = -\frac{1}{2}$, so all electrons will be in the spin-down position. This is consistent with equation 2.11, since then the ratio will be zero. Plotted below in figure 2.4 is the spin temperature T_m versus the average spin s.

A nice thing about the Weiss model is that it predicts a critical point. If we combine equation 2.10 and 2.12 we see after taking the limit $s \to 0$:

$$T_c \equiv \lim_{s \to 0} T_m = \frac{2E_{m0}}{k_B} \lim_{s \to 0} \frac{s}{\ln \frac{\frac{1}{2}+s}{\frac{1}{2}-s}} = \frac{2E_{m0}}{k_B} \cdot \frac{1}{4} = \frac{E_{m0}}{2k_B} \tag{2.13}$$

where T_c is the so called Curie-temperature. So that means there is a finite temperature that corresponds to zero average spin. For Ni, which is commonly used in these experiments, T_c has a value of $620K$. Equation 2.13 also shows that specifying T_c determines E_{m0}.
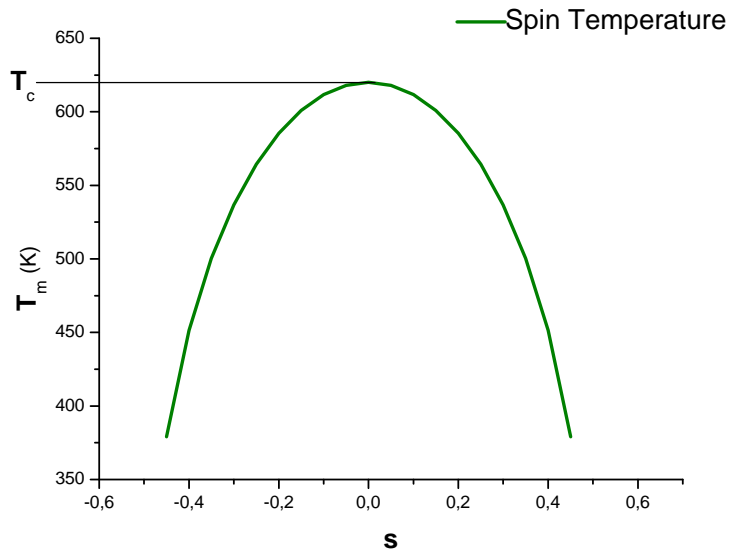
Figure 2.4: Temperature versus magnetization curve.

2.2 Interactions

2.2.1 Boltzmann-equations

The dynamics in our model are mostly described by so called Boltzmann equations. These rate equations count the probabilities of transitions of electrons from one level to another as a result of a particular interaction. Consider for example a transition from the energy level E_i to $E_{i'}$ and vice versa (see figure 2.5 below).
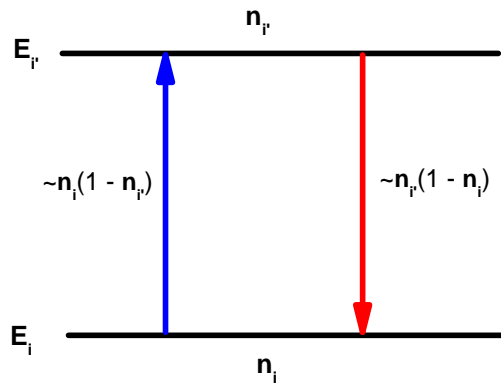
Figure 2.5: Example of a transition between two energy levels.

The general Boltzmann equation describing this process looks like:

$$\frac{dn_i}{dt} = -\frac{dn_{i'}}{dt} = -K \cdot [n_+ n_i(1 - n_{i'}) - n_- n_{i'}(1 - n_i)] \tag{2.14}$$

where K is an interaction constant usually consisting of several factors. There are two terms in equation 2.14, the first representing transitions from i to i' and the second the other way around. Each term contains a factor $n_\pm$, the probability of occurrence of the interaction responsible for the transition, and the combination $n_i(1 - n_{i'})$ or $n_{i'}(1 - n_i)$, which is the probability of finding an electron within the energy range of E_i (represented by n_i) times that of having a free space at $E_{i'}$ (represented by $1 - n_{i'}$) or the other way around in the second case.

2.2.2   Electron-electron scattering

The electron system is internally equilibrated by electron-electron scattering. This process can be viewed at in the following way (see figure 2.6 below).
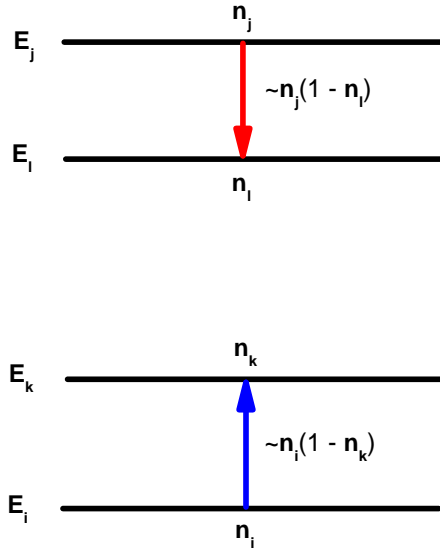
Figure 2.6: Representation of electron-electron scattering.

Two electrons collide with each other and redistribute their total energy. The rate of occurrence of this event is also described by a Boltzmann equation. We use the notations defined in section 2.1.1. If the starting energies are E_i and E_j and the energies after the collision are E_k and E_l then the rate of change is:

$$\frac{dn}{dt} = P_{ee} D_e^2 (\Delta E)^2 n_i n_j (1 - n_k)(1 - n_l) \tag{2.15}$$

where P_{ee} is an interaction constant and E_i, E_j, E_k, E_l are such that the total energy is conserved. It is of course also required that the total number of electrons remains the same, meaning:

$$\frac{dn_i}{dt} = \frac{dn_j}{dt} = -\frac{dn_k}{dt} = -\frac{dn_l}{dt} \tag{2.16}$$

where this is all equal to $-\frac{dn}{dt}$ from equation 2.15.

Because equation 2.15 contains four different indices (i,j,k and l) which are related through the conservation of energy, the calculation of this scattering process involves a three dimensional summation. In the execution of the model this is a very time consuming step, so in most of our simulations we will just assume infinitely fast thermalization.

2.2.3 Laser excitation

At the start of the experiment a laser pulse hits the material and instantly interacts with the electrons. What happens is that some electrons will absorb a photon and thus go to a higher energy state and this will initiate the demagnetization process. In the model this is implemented in the following way. We assume a constant photon energy E_{photon}, so an electron can jump from a state with energy E to one with $E + E_{photon}$. If we look back at the representation of the electron system, this event will most likely happen when there are a lot of electrons with energy E and only very few with energy $E + E_{photon}$ (see figure 2.7 below).
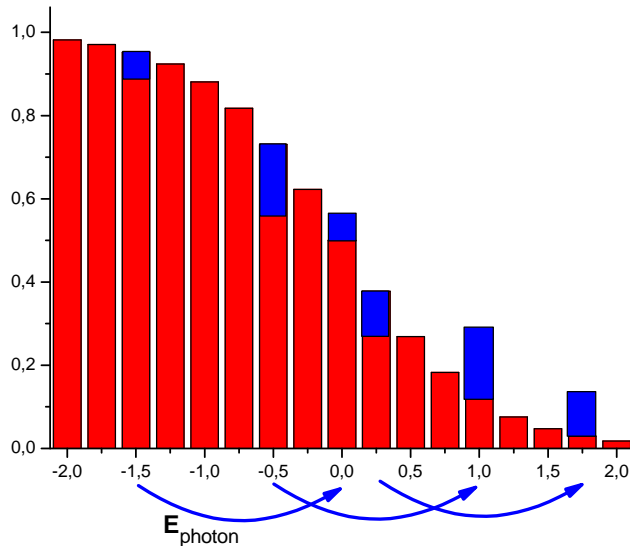
Figure 2.7: Representation of photon absorption by the electrons.

Therefore we take:

$$\Delta n_i = -\Delta n_{i'} = P_l n_i (1 - n_{i'}) \qquad (2.17)$$

where $i' > i$ is such that $E_{i'} = E_i + E_{photon}$, and $P_l < 1$ is a constant that depends on the laser intensity.

As explained earlier, we will not use electron-electron scattering in most of the simulations because it requires a lot of computer time. This means the approach for the laser excitation described here cannot be used because it would put the electron distribution out of equilibrium. Instead we will just model the excitation

as a simple increase of the electron temperature. This won't have any major effect on the demagnetization process, which is what we are interested in.

2.2.4 Electron-phonon scattering

Electron-phonon scattering is the mechanism that equilibrates the electron and lattice systems to each other. An electron with energy E_i interacts with the lattice, and either emits or absorbs a phonon with energy E_p (as defined in section 2.1.2). So the resulting electron will have an energy of $E_i + E_p$ or $E_i - E_p$. We use the general approach of section 2.2.1 by looking at the exchange of electrons between two different energy levels.

Take $i' > i$ such that $E_{i'} = E_i + E_p$, then a phonon absorption corresponds to an electron going from energy E_i to $E_{i'}$, and a phonon emission to the other way around. The rate of occurrence of these two events is described by the following equation:

$$\frac{dn_i}{dt} = -\frac{dn_{i'}}{dt} = -P_{ep}D_e D_p \cdot [n_p n_i(1 - n_{i'}) - (1 + n_p)n_{i'}(1 - n_i)] \qquad (2.18)$$

where P_{ep} is an interaction constant. The $1 + n_p$ term is from phonon emission (it is always possible to emit a phonon) and the n_p term from phonon absorption (there has to be a phonon first to be able to absorb one). The extra energy per unit of time $\frac{dE_{ii'}}{dt}$ that is put into the electron system due to this interaction is:

$$\frac{dE_{ii'}}{dt} = -D_e \Delta E \frac{dn_i}{dt}(i' - i)\Delta E = -D_e \Delta E \frac{dn_i}{dt} E_p \qquad (2.19)$$

where we used equation 2.1 for the total amount of electrons in a cell.

Equation 2.18 describes completely the changes to the electrons by giving the rate of change of n_i for each cell. However, similar to the laser excitation, when we don't include electron-electron scattering as an interaction we have to use a different way. In that case we use the change in energy from equation 2.19 combined with expression 2.6 for the electronic heat capacity c_e to calculate the change in the electron temperature T_e:

$$\frac{dT_e}{dt} = \frac{1}{c_e}\frac{dE_e}{dt} = \sum \frac{1}{c_e}\frac{dE_{ii'}}{dt} = -D_e \Delta E \frac{E_p}{c_e}\sum \frac{dn_i}{dt} \qquad (2.20)$$

We are also interested in what happens to the lattice. Energy is conserved so that means the temperature of the lattice T_l will change a little. Using the same approach as for the electrons we find:

$$\frac{dT_l}{dt} = \frac{1}{c_l}\frac{dE_l}{dt} = \sum -\frac{1}{c_l}\frac{dE_{ii'}}{dt} = D_e \Delta E \frac{E_p}{c_l}\sum \frac{dn_i}{dt} \qquad (2.21)$$

2.2.5 Electron-phonon scattering with spin-flip

Electron-phonon scattering with spin-flip is the interaction that links the other two systems to the spin-system. This mechanism is very similar to the electron-phonon interaction, except that now there is also a small probability α that a phonon absorption/emission is accompanied by a spin-flip of the electron. The resulting electron energy will then be $E_i \pm E_p \pm E_m$, depending on whether it reflects the absorption/emission of a phonon and on the original spin state of the electron.

We use the same approach of looking at the exchange of electrons between the

two energy levels E_i and $E_{i'}$, where we take the energy difference $E_{i'} - E_i$ to be $E_p \pm E_m$. This covers all the four possible events.

The rate of occurrence of these events is described (similarly to equation 2.18) by:

$$\frac{dn_i}{dt} = -\frac{dn_{i'}}{dt} = -\alpha P_{ep} D_e D_p \cdot [n_p(\frac{1}{2} \pm s)n_i(1-n_{i'}) - (1+n_p)(\frac{1}{2} \mp s)n_{i'}(1-n_i)] \quad (2.22)$$

where the factors $(\frac{1}{2} \pm s)$ represent the probability of a transition from the spin-up to the spin-down state or vice versa. Notice that equation 2.18 will now get an extra factor $(1-\alpha)$ in front of it, because we have to make the distinction between spin-flip and non spin-flip events.

Now we also need a way to calculate the changes to the average spin as a result of this interaction. As defined earlier there are a number of D_m spins per atom, so that means each spin-flip changes the average spin by $\pm\frac{1}{D_m}$ (depending on the original spin state). Using again equation 2.1 we get as a result:

$$\frac{ds}{dt} = \sum D_e \Delta E \frac{dn_i}{dt} \frac{\pm 1}{D_m} \quad (2.23)$$

2.2.6 Heat diffusion through lattice

The laser pulse that initiates the experiment causes an amount of excess energy in the system. A way to release this energy is through the lattice. We consider the lattice connected to an external heat bath, which is represented by a similar lattice system (with infinite heat capacity in our case). The interaction between them is described by a simple equation:

$$\frac{d(T_1 - T_2)}{dt} = -\frac{1}{\tau_{12}}(T_1 - T_2) \quad (2.24)$$

where τ_{12} is a time constant. Energy considerations and the use of equation 2.9 for the energy of the lattice give:

$$c_1 T_1 + c_2 T_2 = constant$$
$$c_1 \frac{dT_1}{dt} = -c_2 \frac{dT_2}{dt} \equiv \frac{\Delta E_{12}}{dt}$$
$$\frac{d(T_1 - T_2)}{dt} = (1 + \frac{c_1}{c_2})\frac{dT_1}{dt} = -\frac{1}{\tau_{12}}(T_1 - T_2)$$
$$\frac{\Delta E_{12}}{dt} = -c_1 \frac{1}{1 + \frac{c_1}{c_2}} \frac{T_1 - T_2}{\tau_{12}} = -\frac{T_1 - T_2}{\tau_{12}(\frac{1}{c_1} + \frac{1}{c_2})} \quad (2.25)$$

where ΔE_{12} is the energy difference between the two lattices. This last equation allows us to calculate the changes in the two temperatures T_1 and T_2 easily.
There is one remark about this derivation. The heat capacities c_1 and c_2 are defined per atom, so if the sizes of the two lattices were different we would have to take that into consideration.

2.2.7 External magnetic field

An interesting addition to the model would be to have an external magnetic field. Surprisingly enough it turns out to be very simple to implement this feature. Since

an external field will affect the spin system there is only one equation that has to be changed. That is equation 2.10 for the energy difference $E_m(s)$ between the spin-up and spin-down state:

$$E_m(s) = -2E_{m0}(s + s_{ex}) \tag{2.26}$$

where s_{ex} is the contribution from the external magnetic field.

We can relate s_{ex} to the external magnetic field H_{ex} as follows. The change in energy $\Delta E_m(s)$ due to the external field is equal to $2g\mu_B H_{ex}$, where μ_B is the magnetic moment of a single electron (Bohr magneton) and $g \approx 2$. Combining this with equation 2.26 we find that $2g\mu_B H_{ex} = -2E_{m0}s_{ex}$. This results in:

$$H_{ex} = -\frac{E_{m0}}{g\mu_B}s_{ex} = -\frac{2k_B T_c}{g\mu_B}s_{ex} \approx 922 s_{ex} \tag{2.27}$$

Sofar all the equations for the spin system are symmetric in the spin-up and spin-down state. We chose the spin-down state to have the lowest energy, but that was just a matter of convenience. By introducing an $s_{ex} \neq 0$ this symmetry is broken and the spins will have a preferred direction. This allows for some interesting simulations that will be discussed in section 3.3.

2.3 Validation

2.3.1 Electron thermalization

A first thing that would be interesting to check is whether the electron system behaves like we want it to. Equations 2.15 and 2.16 should describe the internal thermalization towards the equilibrium state given by the Fermi-Dirac distribution 2.3.

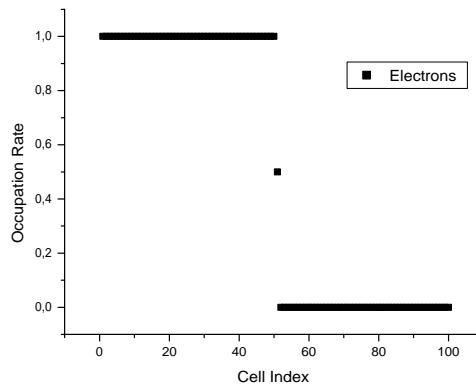If we start out with $T_e = 0$ the electronic states look like this:

Figure 2.8: Distribution of electronic states at zero temperature.

If we now disturb this equilibrium by using a laser excitation for instance, it will change instantaneously:
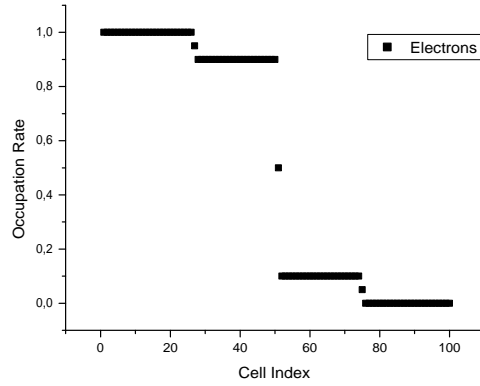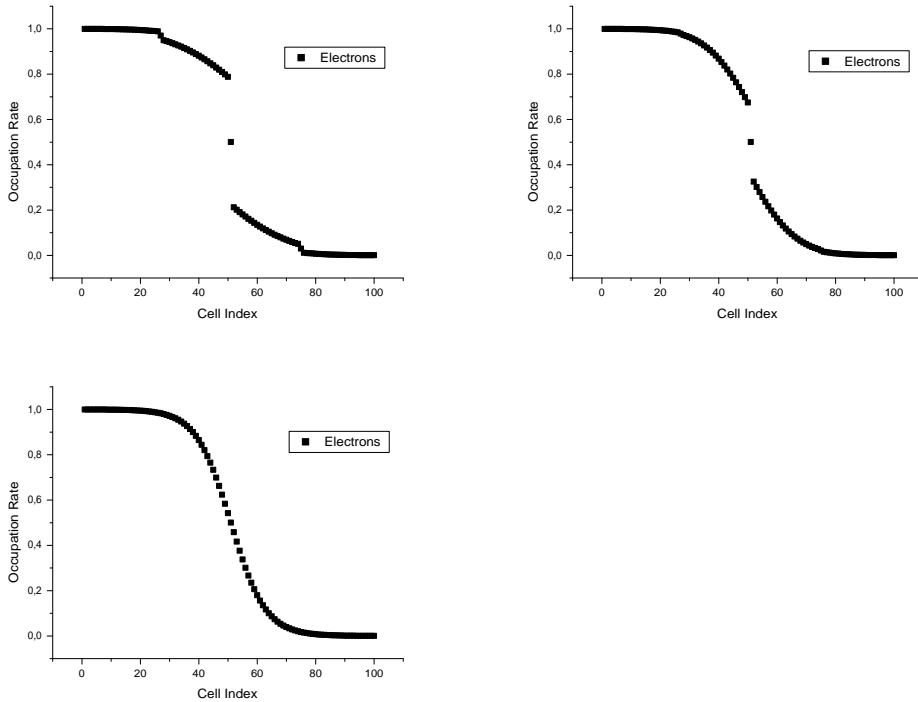

Figure 2.9: Distribution of electronic states right after a laser excitation.

The following graphs show how over a certain time it gradually changes into a smooth Fermi-Dirac distribution with a temperature $T_e > 0$.

This demonstrates that the equations we used are indeed a good way to describe the dynamics of the electron system.

2.3.2 Temperature equilibrium

When there are several subsystems that can interact with each other eventually they will reach some kind of equilibrium state. In that situation you would expect them to all have the same temperature. The following graphs show that this behavior is indeed what our model predicts.
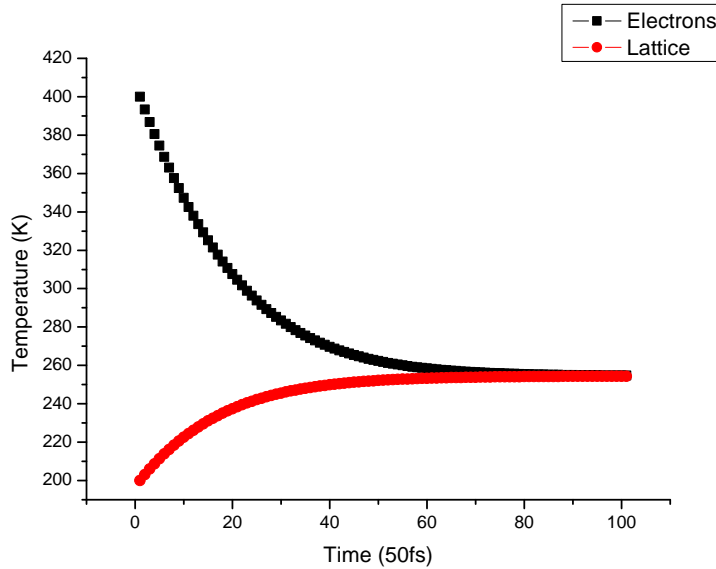
Figure 2.10: Electron-lattice temperature equilibrium.

This graph shows the equilibration of the lattice and the electrons. At $t = 0$ the temperature of the two subsystems are set to different values ($200K$ and $400K$ in this case). The system then evolves according to the electron-phonon scattering equations described in subsection 2.2.4. After a while they reach an equilibrium state and both temperatures are equal to each other ($T_e = T_l \approx 255K$ in this example). The final temperature depends of course upon the specific parameter values of D_e, c_l etc. The other two graphs below are simulations of a similar experiment, but now for different systems.

Figure 2.11: Electron-lattice-spin temperature equilibrium.

Figure 2.12: Lattice lattice temperature equilibrium.

Chapter 3
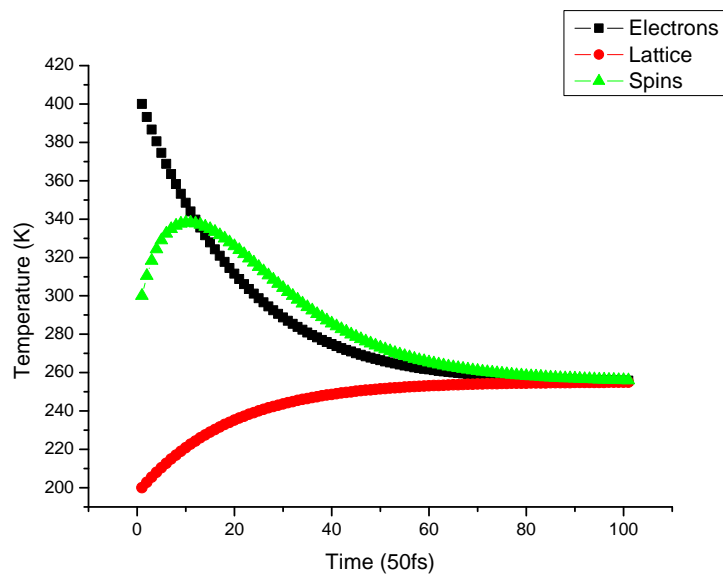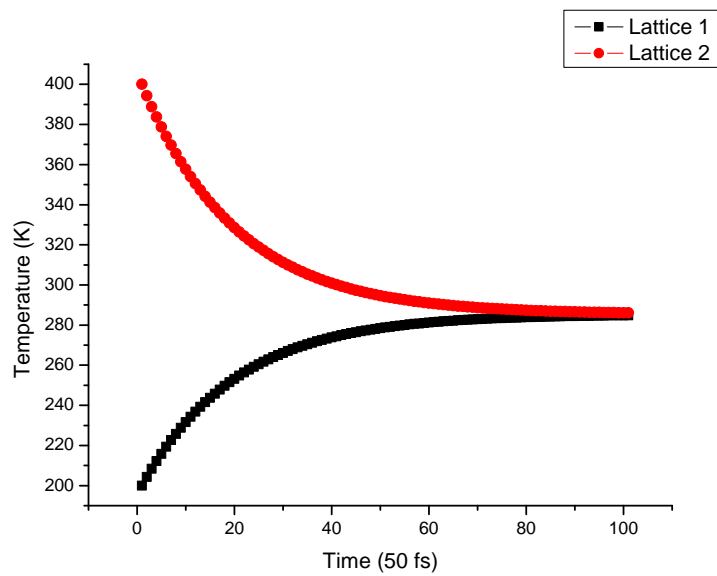
Results

Now that we have a flexible and working model capable of simulating the experiment described in chapter 1, it is interesting to see how the system behaves as a function of certain parameters. These will be discussed in the next sections.

3.1 α-dependence

The first and most interesting parameter to check is the spin-flip probability α. It was defined as the relative number of electron-phonon scattering events that are accompanied by a spin-flip. It can be expected that the response of the spin system depends a lot on this parameter. In the literature it is often claimed that a phonon based mechanism, such as we have proposed in the model, is necessarily slower than the electron-phonon equilibration. We want to see if there are possible values for α that allow for a demagnetization that goes faster than this equilibration.

The sort of simulation that we will be using for this is the following. We have the tree subsystems electrons, lattice and spins. We let the electrons start out at a temperature of $400K$ while the other two systems have an initial temperature of $300K$. We then let the systems evolve according to the equations described in subsections 2.2.4 and 2.2.5. It should be noted that we assume infinitely fast electron thermalization, so we have to use the alternative equation 2.20 to describe the changes to the electron system. Plotted below are the results from these simulations.
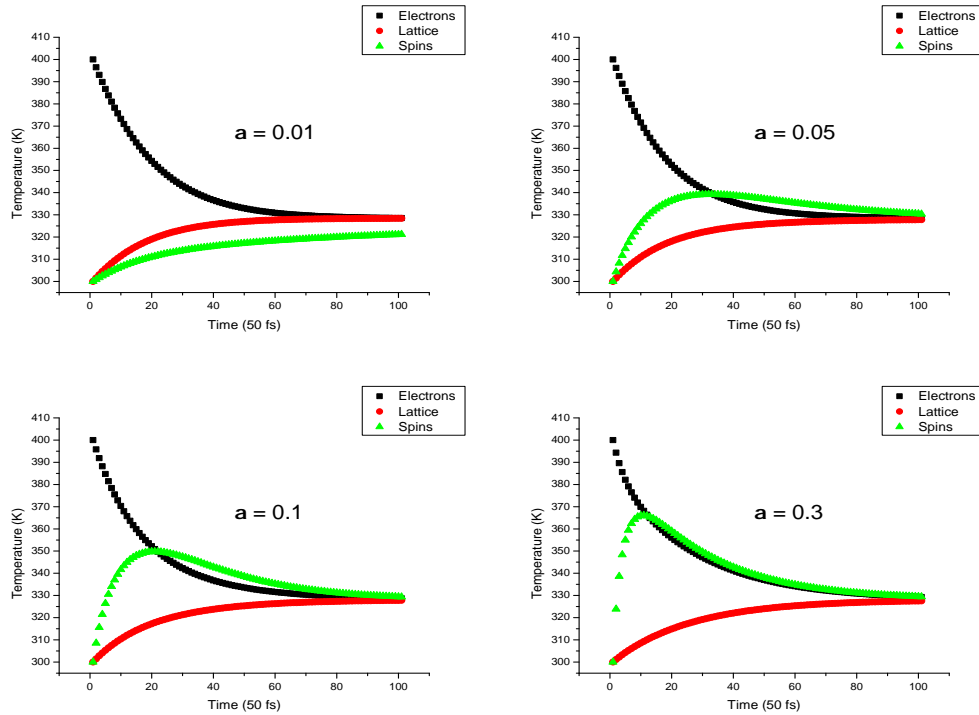
As we can see for different values of α the spin dynamics change completely. In the graph with $\alpha = 0.01$ the spin temperature curve displays an exponential growth that is slower than the e-p equilibration process. For $\alpha \geq 0.05$ the response of the spin temperature is faster, showing a double exponential behavior. As it turns out these values for α are not unrealistic for Ni (see Kicken[1]). This demonstrates that the demagnetization can indeed be faster than electron-phonon equilibration with a phonon mediated spin-flip mechanism.

3.2   Verification of τ_e

In this section we ignore the spin system for a moment and only look at the equilibration of the electrons and the lattice. In the model this corresponds to setting α equal to zero. An expression for the timescale τ_e of this process can be derived (see Koopmans[2]). In first approximation the result is:

$$\tau_e = \frac{\hbar\pi k_B T_e}{6 D_e D_p E_p \lambda_{ep}^2} \tag{3.1}$$

where λ_{ep} is a constant independent of the rest. By varying the model parameters D_e, D_p and E_p we will show that our model agrees with equation 3.1.

A typical graph of this experiment is shown in fig 3.1.

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[1]H.H.J.E. Kicken. *Report, Ultrafast magnetization dynamics in ferromagnetic materials, Eindhoven University of Technology, The Netherlands*, 2005.

[2]Prof. Dr. B. Koopmans, *Comparison of Ultrafast demagnetization, Gilbert damping and e-p relaxation, Eindhoven University of Technology, Netherlands*, 2005.
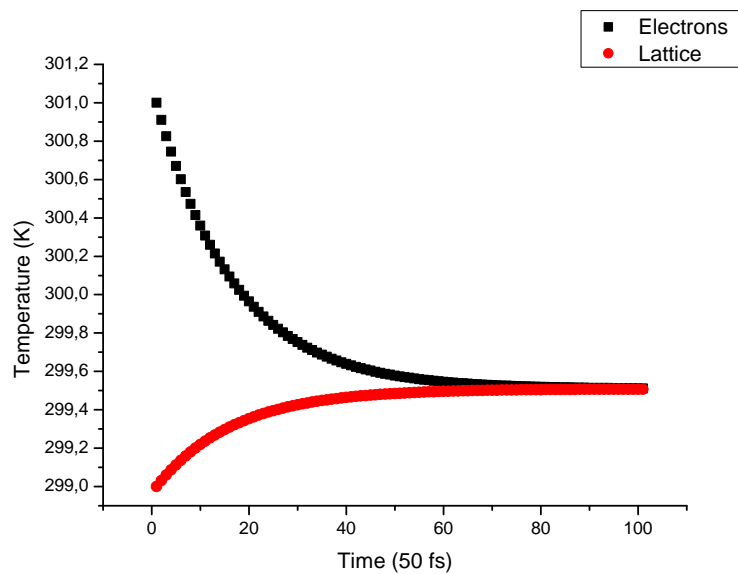
Figure 3.1: Electron-phonon relaxation example.

The electron temperature curve can be fitted with an exponential decay yielding an estimate for the relaxation time τ_e. By repeating the simulation for different values of D_e, D_p and E_p while using the same fitting procedure we obtain the following three graphs of τ_e.
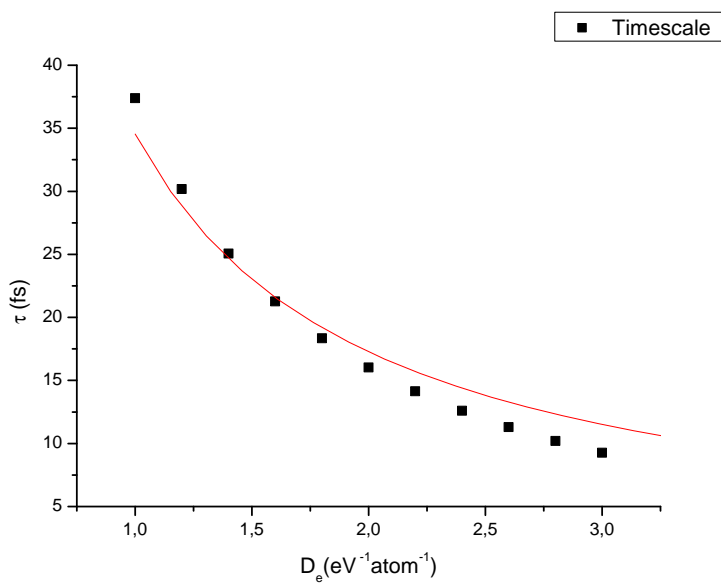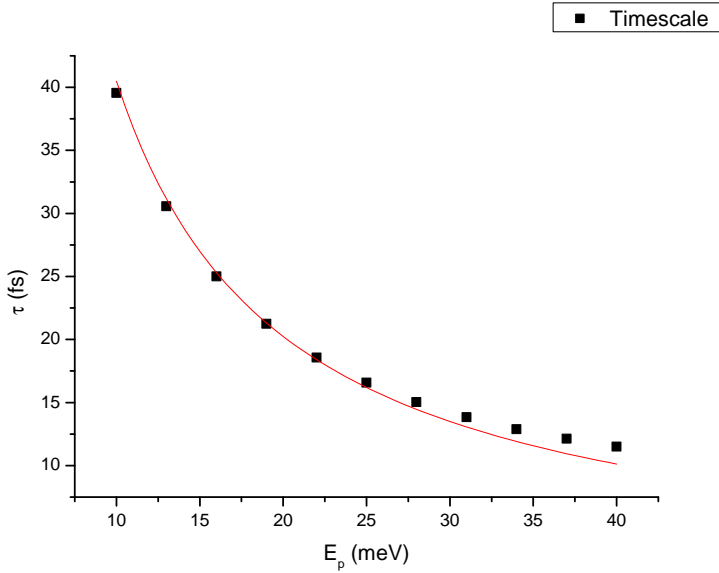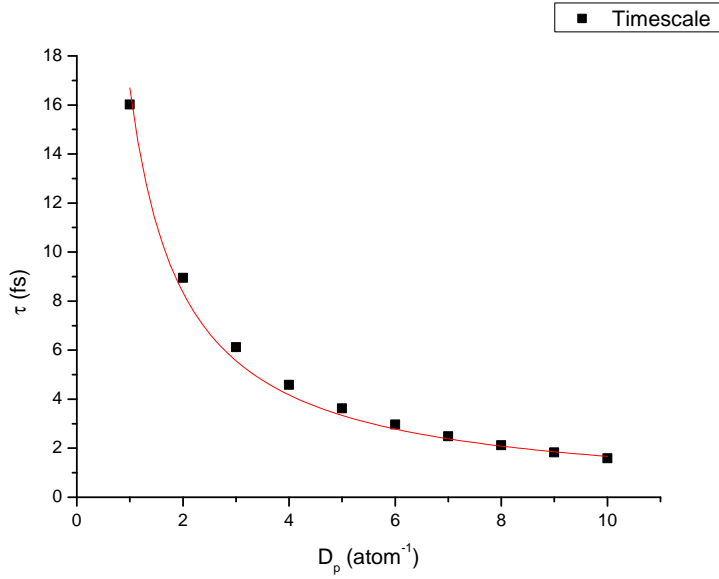
In all graphs we have plotted the $\frac{1}{x}$-fit (displayed as the red line) to illustrate the behavior of the curve. It can be seen that this fit corresponds nicely to the data in all three cases. We conclude that in a first approximation the timescale τ_e does indeed vary with the inverse of D_e, D_p and E_p.

3.3   Magnetic switching

The addition of an external magnetic field in combination with the possibility of heat diffusion through the lattice makes it possible to simulate magnetic switching experiments. To illustrate this we first consider a simulation similar to the one described in section 3.1, except that we now increase the laser power to speed up

the demagnetization process. Effectively this means the electron temperature T_e is instantaneously raised to $850K$ instead of $400K$. The result is displayed in figure 3.2 below.
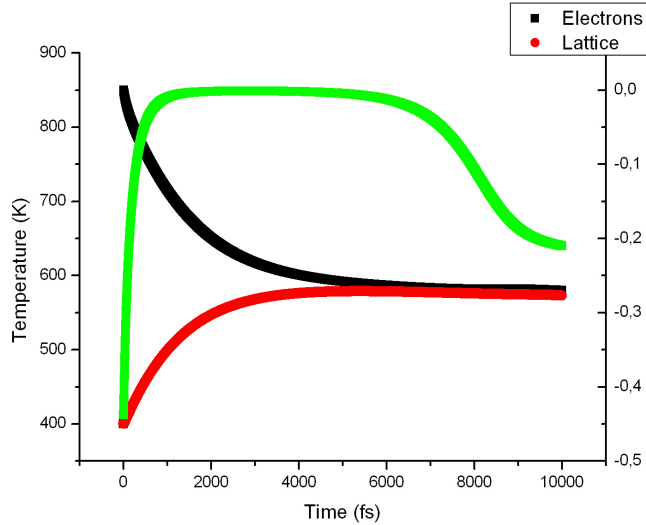
Figure 3.2: Example of a demagnetization experiment with high laser power.

Instead of the spin temperature T_m we show the behavior of the average spin s by the green line (the scale is displayed on the right side). The reason for this is that there is no definition of T_m for temperatures above the Curie-temperature T_c. It can be seen that the response of the spin-system is very fast (we took $\alpha = 0.3$) but s never exceeds the zero-line. This is because when s approaches zero the energy difference between the spin-up and spin-down state also goes to zero. That means there will very likely be just as many spin-flips in one direction as in the other and it becomes increasingly more difficult to increase the average spin further. After a while, when the electrons and the lattice temperature have stabilized, the spin-system can release its excess energy again and equilibrate with the rest.

If we now apply a small external field by setting $s_{ex} = 0.0001$ in equation 2.26 we get a completely different graph, shown in figure 3.3 below.
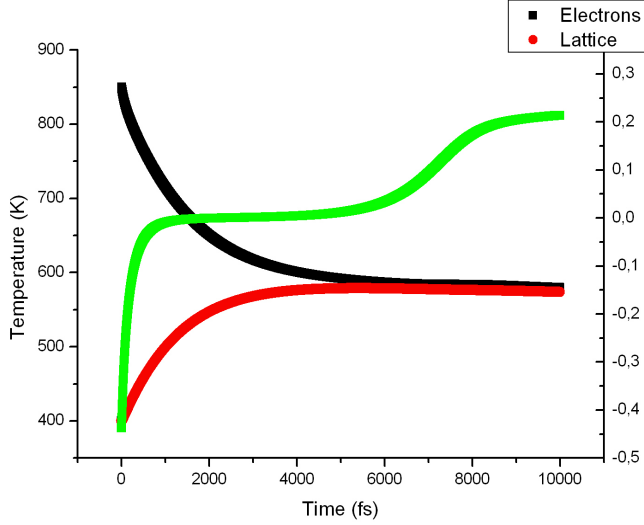
Figure 3.3: High laser power and a small magnetic field.

In the start the graphs look alike, but at around $5000fs$ the real difference is clearly visible. The average spin now goes through zero and grows back into the other direction. This can be understood by recognizing that $E_m(s)$ changes sign when $|s|$ becomes smaller than s_{ex}. At that point the spin-up state will have a lower energy than the spin-down state, which means the roles are reversed and the spin-system can only reach equilibrium again by growing back in the positive direction. This shows that by using a laser excitation and applying a small external field we can enforce the magnetization to switch sign.

For this magnetic switching to occur it is necessary that the spin-system reaches a high enough temperature, otherwise s will not exceed $-s_{ex}$. Figure 3.4 shows what happens when the laser power is slightly decreased. The average spin grows back in the original direction and no magnetic switching occurs.
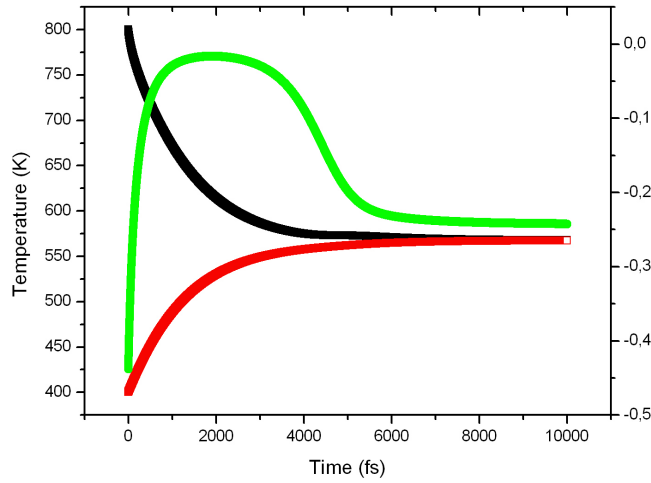
Figure 3.4: Slightly lower laser power and a small magnetic field.

To speed up the remagnetization process we can use cooling by an external heat bath (see figure 3.5). The temperature of the heat bath is fixed at $400K$, the starting temperature of the lattice and spins.
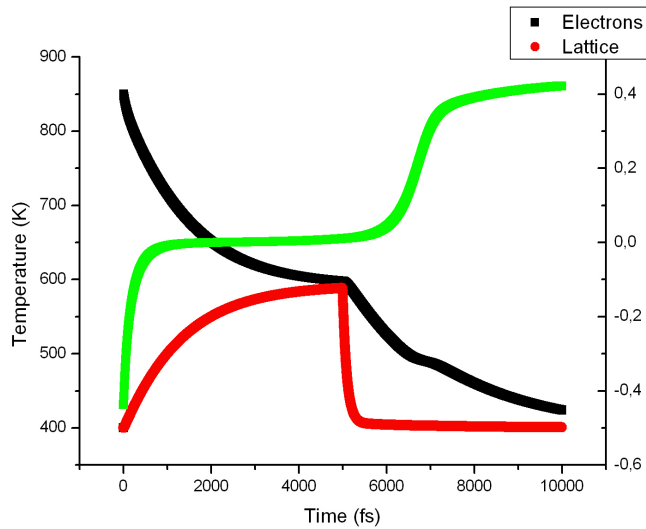
Figure 3.5: Magnetic switching with strong cooling enabled at $5000fs$.

At $t = 5000fs$ the lattice starts to cool down very quickly as a result of the heat diffusion. The electrons and the spin-system follow this behavior. As can be seen, the remagnetization occurs much more rapidly now.

Chapter 4

Conclusions

In this report we have derived a microscopic model for the ultra fast demagnetization that occurs after heating a ferromagnetic material with a short laser pulse. We assumed phonon-mediated spin-flip scattering to be the main cause for the demagnetization. We defined a spin-flip probability α for every electron-phonon scattering event. In contradiction to claims in literature, it was shown that phonon-mediated demagnetization can be faster than electron-phonon equilibration. Furthermore we have investigated the possibility of magnetic switching by heating the spin-system above the Curie temperature and applying an external magnetic field.

Appendix A

Code Example

What follows in this section is an example of the code used to simulate one of the experiments described in this report. The model has been programmed in a highly object-oriented fashion such that only a very limited amount of knowledge about the internal structure of the underlying objects is needed to be able to use the program. Everything was made in C++ with Borland's CBuilder 6, therefor it is also needed to be able to run it.

The one important file that contains all the information about what type of simulation you are running and what are the parameter values is *Unit1.cpp*:

```
//---------------------------------------------------------------------------
#include <vcl.h>
#pragma hdrstop
#include "Model.h"
#include "SimpleElectrons.h"
#include "WeissSystem.h"
//---------------------------------------------------------------------------

#pragma argsused

int main(int argc, char* argv[])
{
 TModel* Model1=new TModel;
 Model1->TimeStep = 1.0;
 Model1->NumberOfSteps = 10000;
```

First we create the Model-object called *Model1*, which takes care of the execution and the output of a simulation. It has two important properties *TimeStep* and *NumberOfSteps* that can be changed. What follows next is a list of subsystems and interactions that we put into the model. It is perhaps useful to know that all subsystem and interaction objects are descendants from their respective base objects *TSubSystem* and *TInteraction*.

```
 TSimpleElectronSystem* ElecSys = new TSimpleElectronSystem;
 Model1->SubSystemList->Add(ElecSys);
 ElecSys->NumberOfCells = 2000;
 ElecSys->DensityOf = 2.0;
 ElecSys->EnergyWidth = 0.0005;
 ElecSys->Temperature = 300.0;
```

Here we create an electron system named *ElecSys* and add it to the list of subsystems of the Model-object. At the same time we set several of its properties.

```
TSimpleLaserExcitation* LaserInteraction = new TSimpleLaserExcitation;
LaserInteraction->Parent = Model1;
LaserInteraction->ElecSys1 = ElecSys;
LaserInteraction->TimeDelay = 10;
LaserInteraction->LaserTemperatureIncrease = 250.0;
```

This is an example of how interactions are incorporated in the model. We assign *Model1* to its *Parent* property, which automatically adds the interaction to the list of interactions in the Model-object as well as making sure that the interaction object knows what Model-object specifically it is being added to. We then tell it what subsystems it should work on. In this case, because it is a laser interaction that only works on one electron system, we set the property *ElecSys1* to be the electron system *ElecSys* that we created earlier on.

```
TPhononSystem* PhononSys = new TPhononSystem;
Model1->SubSystemList->Add(PhononSys);
PhononSys->HeatCapacity = 4.31E-5;
PhononSys->PhononEnergy = 0.026;
PhononSys->DensityOf = 1.0;
PhononSys->Temperature = 300.0;

TPhononSystem* PhononSys2 = new TPhononSystem;
Model1->SubSystemList->Add(PhononSys2);
PhononSys2->Infinite = True;
PhononSys2->Temperature = 300.0;
```

The property *Infinite* is used if you want subsystems that are of infinite size. In this case it means the heat capacity of the phonon system is infinite.

```
TWeissSpinSystem* SpinSys = new TWeissSpinSystem;
Model1->SubSystemList->Add(SpinSys);
SpinSys->CurieTemperature = 620.0;
SpinSys->SpinFlipProbability = 0.1;
SpinSys->ExternalField = 0.0001;
SpinSys->DensityOf = 0.08;
SpinSys->Temperature = 300.0;

TSimpleElectronPhononSpinInteraction* Interaction =
    new TSimpleElectronPhononSpinInteraction;
Interaction->Parent = Model1;
Interaction->ElecSys1 = ElecSys;
Interaction->PhononSys1 = PhononSys;
Interaction->SpinSys1 = SpinSys;
```

This interaction works on multiple subsystems at the same time, so it needs to know all of them to function properly. Note that we used *PhononSys* here and not *PhononSys2*, which means we are including the lattice with finite heat capacity.

```
TPhononPhononInteraction* Interaction2 = new TPhononPhononInteraction;
Interaction2->Parent = Model1;
Interaction2->TimeDelay = 5000;
Interaction2->TimeScale = 1000.0;
```

```
 Interaction2->PhononSys1 = PhononSys;
 Interaction2->PhononSys2 = PhononSys2;

 Model1->Execute();
```

We start the simulation by calling the *Execute* function of the Model-object.

```
 delete Model1;
 return 0;
}
//------------------------------------------------------------------------
```

After the program is done running, several output files will be saved to the *Output* directory in the folder where the original program files were placed.

Appendix B

A system of electrons with discrete energy levels

B.1 Assumptions and definitions

We consider a (large) number N of electrons which can occupy a discrete set of quantum states. To keep things simple we assume the energy levels of the states are evenly spaced with distance 1, and each distinct energy gives room to the same finite amount of states M. To keep all the relevant quantities finite the energy levels run from $-L$ to L, but our main interest lies in the limit of $L \to \infty$. At zero temperature we assume that all states up to energy 0 are completely filled and all the others empty. In essence this means that (for our own convenience) we take the Fermi level to be at zero energy. Note that this implies that N is equal to half of the total number of available states $(2L+1) * M$. We don't really care about which exact states are occupied, only the amount of states that are occupied at a certain energy is important. And so we define the numbers n_i as the relative amount of occupation at the energy i, which is the number of occupied states divided by M, where $i \in \mathbb{Z}$ and $0 \le n_i \le 1$. If we plot these numbers n_i at zero temperature we get the distribution D_0 (where we took $L = 10$) like in figure B.1.
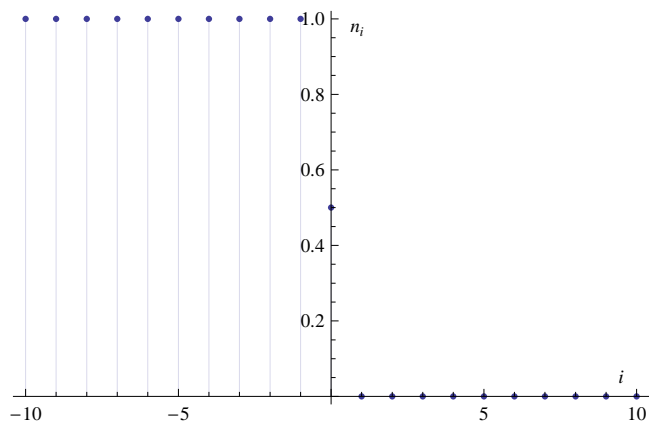

Figure B.1: The distribution of states at zero temperature.

B.2 Energy

For a distribution of the n_i we define an **energy** in the following way. To the distribution at the absolute zero D_0 (see figure B.1) we assign an energy of zero. For all other distributions we take the energy to be the amount of additional energy that is in the system relative to D_0, again divided by the number of states M at each energy. In terms of the n_i this means:

$$E = \sum_{i=1}^{L} i n_i - \sum_{i=-L}^{-1} i(1 - n_i) \tag{B.1}$$

Notice that the term with n_0 is absent since those electrons have zero energy (Fermi level). As an example, the distribution in figure B.2:
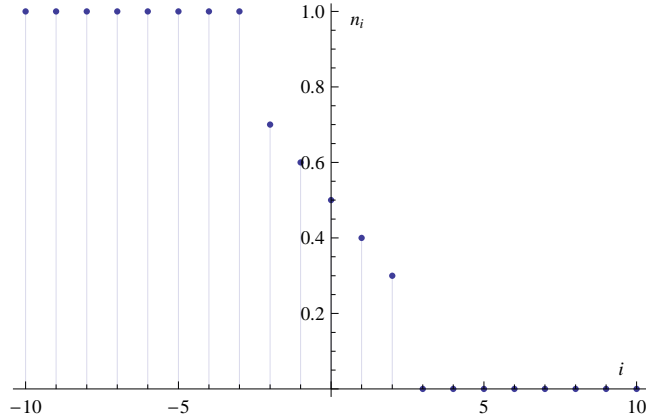

Figure B.2: Example of a distribution with non zero energy.

has an energy of $E = 0.4 + 2 * 0.3 - (-1) * (1 - 0.6) - (-2) * (1 - 0.7) = 2$.

B.3 Electron scattering

The principal process that is responsible for the evolution of the n_i is the scattering of two electrons. This process is depicted in figure B.3 below.

An electron of energy i collides with an electron of energy j. A certain amount of energy is transferred between them in the collision and the first electron goes to a free state of energy k and the second to one of energy l. The total amount of energy is of course conserved and so it has to hold that:

$$i + j = k + l \tag{B.2}$$

We assume that the relative rate $r(i, j, k, l)$ at which this process occurs, that is the number of scattering events of the type above that occur each second divided by M, satisfies the following simple relation:

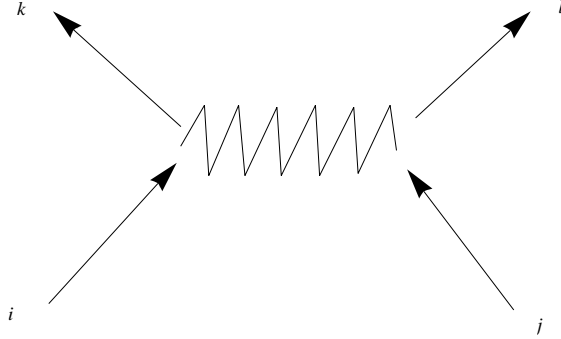$$r(i, j, k, l) = \frac{1}{L^2} n_i n_j (1 - n_k)(1 - n_l) \tag{B.3}$$

Figure B.3: Graphical representation of the scattering of two electrons.

where i, j, k, l satisfy B.2. The intuition behind this equation is that in order to have many scattering events there need to be a lot of electrons at energies i, j and a lot of free states for them to go to at k, l. The factor $\frac{1}{L^2}$ is there to account for the scaling.

The reverse process, in which two electrons of energies k, l collide and go to a states of energy i, j, occurs with the rate $r(k, l, i, j) = \frac{1}{L^2} n_k n_l (1 - n_i)(1 - n_j)$. The net rate of change $R(i) := \frac{\partial n_i}{\partial t}$ of the relative occupation at energy i can be acquired by summing the two rates above (with appropriate sign) over all possible j, k, l:

$$R(i) = \frac{1}{L^2} \sum_{\substack{j,k,l \\ i+j=k+l}} [r(k, l, i, j) - r(i, j, k, l)] \tag{B.4}$$

B.4 Equilibrium

B.4.1 General solution

The distributions of the n_i which are in equilibrium are those that do not change over time, which means:

$$R(i) = \frac{1}{L^2} \sum_{\substack{j,k,l \\ i+j=k+l}} [r(k, l, i, j) - r(i, j, k, l)] = 0, \quad \forall i \tag{B.5}$$

Let us try to find a solution for which each term in this sum is equal to zero. We have:

$$n_i n_j (1 - n_k)(1 - n_l) = n_k n_l (1 - n_i)(1 - n_j)$$
$$\frac{n_i}{(1 - n_i)} \frac{n_j}{(1 - n_j)} = \frac{n_k}{(1 - n_k)} \frac{n_l}{(1 - n_l)}$$

If we rename $\beta_i := \frac{n_i}{(1-n_i)}$ this last equation becomes $\beta_i \beta_j = \beta_k \beta_l$. Now we fill in the values $i = 0$ and $k = 1$. The constraint on the numbers i, j, k, l says that $0 + j = 1 + l$ which means $j = l + 1$.

$$\beta_0 \beta_j = \beta_1 \beta_l$$

$$\frac{\beta_{l+1}}{\beta_l} = \frac{\beta_1}{\beta_0}$$

This last equation has to hold for all l. If we define $b := \frac{\beta_1}{\beta_0}$ we arrive at the general solution for the β_i:

$$\beta_i = c * b^i \tag{B.6}$$

By definition we want the value of $\beta_0 = 1$, since that means $n_0 = \frac{1}{2}$ which will be consistent with our constraint on the number of electrons. This corresponds to the value $c = 1$ and $\beta_i = b^i$. The general solution $D(a)$ for the n_i now easily follows:

$$n_i = \frac{\beta_i}{1 + \beta_i} = \frac{b^i}{1 + b^i} = \frac{1}{(\frac{1}{b})^i + 1} = \frac{1}{a^i + 1} \tag{B.7}$$

where $a := \frac{1}{b}$. An example of this distribution is shown in figure B.4.
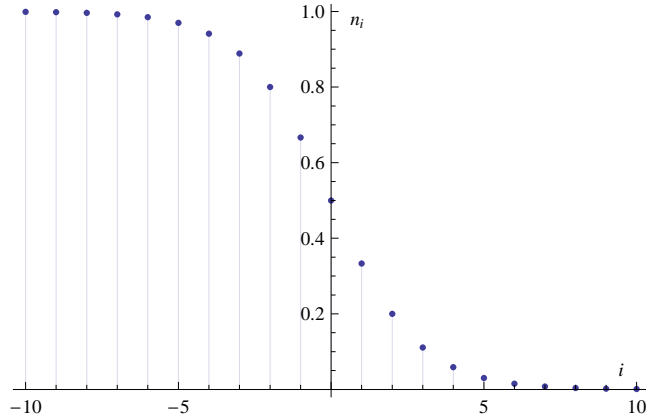
Figure B.4: Plot of the equilibrium distribution for $a = 2$.

Noting that for $D(a)$ it holds that $n_{-i} = \frac{1}{\frac{1}{a^i} + 1} = \frac{a^i}{1 + a^i} = 1 - n_i$ we see that the distribution also satisfies the constraint on the number of electrons in the sense that exactly one half of the states are occupied. In the limit $L \to \infty$ we have $\lim_{L \to \infty} \left(\frac{1}{2L} \sum_{i=-L}^{L} n_i \right) = \frac{1}{2}$.

B.4.2 Entropy

To prove that this is the only possible solution we will use the notion of **entropy**. We define the entropy S_i of state i to be:

$$S_i := - \{ n_i \ln n_i + (1 - n_i) \ln(1 - n_i) \} \tag{B.8}$$

This is similar to the information entropy of a simple Bernoulli random variable with probability $p = n_i$. The total entropy S of the system is now just the sum of S_i over all i, i.e. $S = \sum_{i=-L}^{L} S_i$ which is a function of all the n_i. Hence for any distribution of the n_i which do not change over time the same will hold for S. The time derivative of S will therefore be equal to zero.

Let us compute the time derivative of S_i:

$$\frac{dS_i}{dt} = \frac{\partial S_i}{\partial n_i}\frac{\partial n_i}{\partial t} = -\left\{n_i \cdot \frac{1}{n_i} + \ln n_i + (1 - n_i) \cdot \frac{1}{(1 - n_i)} \cdot -1 - \ln(1 - n_i)\right\}\frac{\partial n_i}{\partial t}$$

$$= -\ln\frac{n_i}{(1 - n_i)} \cdot \frac{\partial n_i}{\partial t} = -\ln\beta_i \cdot \frac{\partial n_i}{\partial t} \tag{B.9}$$

Now suppose that there are i, j, k, l, with $i + j = k + l$, such that $r(i, j, k, l) - r(k, l, i, j) \neq 0$. We define $r := r(i, j, k, l) - r(k, l, i, j)$ and assume that $r > 0$, otherwise we can just switch the roles of i, k and j, l. That means that there is a net flow of electrons from i to k and from j to l due to this interaction. In terms of the β variables the assumption $r > 0$ can be written as:

$$\beta_i\beta_j > \beta_k\beta_l \tag{B.10}$$

Let us now look at the net rate of change in entropy of these four states. We have:

$$r = \frac{\partial n_k}{\partial t} = \frac{\partial n_l}{\partial t} = -\frac{\partial n_i}{\partial t} = -\frac{\partial n_j}{\partial t}$$

$$\frac{d(S_i + S_j + S_k + S_l)}{dt} = r(\ln\beta_i + \ln\beta_j - \ln\beta_k - \ln\beta_l) = r\ln\frac{\beta_i\beta_j}{\beta_k\beta_l} \tag{B.11}$$

where we have used equation B.9 for the time derivative of the state entropy. Now from equation B.10 we know $\frac{\beta_i\beta_j}{\beta_k\beta_l} > 1$ and also $r > 0$ so we conclude $\frac{d(S_i+S_j+S_k+S_l)}{dt} > 0$. For each i, j, k, l like above we see that the total entropy only increases in time. But that implies $\frac{dS}{dt} > 0$ which is in contradiction with the fact that the n_i did not change over time. And hence there cannot exist such i, j, k, l which satisfy our assumption. Notice that if we had assumed $r < 0$ the fraction $\frac{\beta_i\beta_j}{\beta_k\beta_l}$ inside the logarithm would be smaller than 1 and we would have come to the same conclusion.

We have shown that any equilibrium distribution necessarily satisfies $r(i, j, k, l) - r(k, l, i, j) = 0$ for all i, j, k, l such that $i + j = k + l$. This means that the equilibrium distribution from equation B.7, which we derived based on this assumption, is unique. All other distributions have an entropy $S(t)$ which strictly increases in time.

B.4.3 Stability

We have derived a solution, have shown that it is the only possible solution, but we have yet to show that it is also a stable solution. To prove this we require the notion of a **Lyapunov function**. This is a function $V(x) : \mathbb{R}^n \to \mathbb{R}$ with the following two properties:

- $V(x) \geq 0$ with equality if and only if $x = 0$

- $\frac{dV(x(t))}{dt} \leq 0$ with equality if and only if $x = 0$

where $x \in \mathbb{R}^n$ is a function of the time $t \in \mathbb{R}$. The **Lyapunov second theorem on stability** says that if we can find such a function V then the system is asymptotically stable with the equilibrium being $x = 0$.

In our case the vector x is the distribution of the n_i and hence lives in the space $\mathbb{R}^{2L+1}$. Now the question is what should we take for $V(x)$. After some thought we see that the second property of V is very similar to what we have just proven for the entropy function $S(x)$, namely $\frac{dS(x(t))}{dt} \geq 0$ with equality if and only if $x = x^* := D(a)$. So let us define $V(x)$ as:

$$V(x) := S(x^*) - S(x + x^*) \tag{B.12}$$

then we have $\frac{dV(x(t))}{dt} = -\frac{dS(x(t)+x^*)}{dt} \leq 0$ with equality if and only if $x + x^* = x^*$ which means $x = 0$, and the second requirement of a Lyapunov function is met.

To prove the first requirement we will use the method of Lagrange multipliers to derive x^* as the distribution that maximizes the entropy $S(x)$ under the constraints of particle and energy conservation. These constraints are:

- $\frac{1}{2L+1} \sum_i x_i = \frac{1}{2}$ to preserve the number of electrons

- $\sum_{i=1}^{L} i x_i - \sum_{i=-L}^{-1} (1 - x_i) = \sum_i i x_i + \frac{1}{2} L(L+1) = E$ to conserve energy

The Lagrangian function G is:

$$G(x, \alpha, \beta) = S(x) + \alpha \left(\sum_i x_i - \frac{1}{2}(2L+1) \right) + \beta \left(\sum_i i x_i + \frac{1}{2} L(L+1) - E \right)$$

Let us compute the partial derivative of G with respect to x_i:

$$\frac{\partial G}{\partial x_i} = \frac{\partial S_i}{\partial x_i} + \alpha + \beta i = -\ln \left(\frac{x_i}{1 - x_i} \right) + \alpha + \beta i = 0$$

where the derivative $\frac{\partial S_i}{\partial x_i}$ was previously computed in equation B.9. Solving for x_i we get:

$$x_i = \frac{e^{\alpha + \beta i}}{1 + e^{\alpha + \beta i}} = \frac{1}{e^{-\alpha} e^{-\beta i} + 1}$$

The particle constraint determines the value of α, which is zero. This can be seen by noting that for $\alpha = 0$ it holds that $x_{-i} = 1 - x_i$ thus satisfying the constraint, and that by increasing/decreasing α all of the x_i will increase/decrease as well.

The value of β is now completely determined by the energy constraint. If we rename $a := e^{-\beta}$ then we end up exactly at our equilibrium distribution $x_i^* = \frac{1}{1+a^i}$. Hence x^* is not only a distribution with maximum entropy, but also the only possible distribution with this property.

We have now shown that $S(x) \leq S(x^*)$, or similarly $0 \leq S(x^*) - S(x + x^*) = V(x)$ for any distribution x under the given constraints, with equality if and only if $x = 0$. This is precisely the first requirement of a Lyapunov function.

With both requirements satisfied for $V(x)$, the theorem now tells us that x^* is an asymptotically stable solution. This means that any distribution that starts close enough to x^* will converge in time to x^*.

B.4.4 Additional properties

We will now look at some additional properties of the equilibrium distribution in the limit of $L \to \infty$. Because we're interested in the distributions which have a finite energy there is a restriction on the number a. We require that $\lim_{i \to +\infty} n_i = 0$ and $\lim_{i \to -\infty} n_i = 1$. It's not hard to see that this means $a > 1$.

We try to compute the energy $E(a)$ of the distribution $D(a)$:

$$E(a) = \sum_{i=1}^{\infty} i \frac{1}{a^i + 1} - \sum_{i=-\infty}^{1} i \frac{1}{a^{-i} + 1} = 2 \sum_{i=1}^{\infty} \frac{i}{a^i + 1} \tag{B.13}$$

which, since $a > 1$, can be bounded from above by:

$$E(a) \leq 2 \sum_{i=1}^{\infty} \frac{i}{a^i} = 2a * -\frac{\partial}{\partial a} \frac{a}{a-1} = \frac{2a}{(a-1)^2}$$

In the limit $a \to \infty$ we see that $E(a)$ has to go to zero. The distribution that corresponds with it is:

$$\lim_{a \to \infty} \frac{1}{a^i + 1} = \begin{cases} \frac{1}{1+1} = \frac{1}{2} & \text{if } i = 0 \\ \frac{1}{\frac{1}{\infty}+1} = \frac{1}{0+1} = 1 & \text{if } i < 0 \\ \frac{1}{\infty+1} = \frac{1}{\infty} = 0 & \text{if } i > 0 \end{cases}$$

which is exactly the distribution D_0 at zero temperature with zero energy. Similarly the limit $a \to 1$ yields the constant distribution $n_i = \frac{1}{2}$ which has infinite energy (and temperature). The parameter a is completely determined by the temperature. If we rewrite $a = e^{\frac{1}{T}}$ we end up with the familiar Fermi-Dirac distribution:

$$n_i = \frac{1}{e^{\frac{i}{T}} + 1} \tag{B.14}$$