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Effect of Temperature on Physical Properties for Nickel Oxide (NiO) by Using Ising Simulation Model

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Abstract: The main objective of this work to investigate the effect of changing in a factor of temperature on physical properties such as the critical temperature (Tc), the magnetization per spin (M), the energy per spin (E), the magnetic susceptibility (χ), and the specific heat (Cv) of NiO for a 6X6 square lattice in the absence of external magnetic field by using Ising simulation model. The simulation results showed that the magnetization per spin changed from a positive value to a negative value at critical temperature (Tc≈ 47.2679045 J/KB) this indicates that the material transited from Antiferromagnetic to diamagnetic state. Also, the ground state energy of NiO was determined to be -38.125 meV.

Keywords: Square Lattice, Antiferromagnetic, Critical Temperature

1. Introduction

Nickel oxide (NiO) was discovered in 1858 which only found pure at the natural in a few locations around the world. The bulk mineral has a deep green color while nanoparticles of NiO are black color by Bahl, C. R. H., 2006. Bulk NiO has a face centered cubic (fcc) NaCl structure with the space group Fm3m, and a lattice parameter of a = 4.177°A. There are two components of spin configurations due to the non-local exchange interaction. For the first component, the direct exchange interaction between the nearest neighbors of Ni ions favors paring of spins to lower energy. For another one, a very strong interaction comes from the super exchange between the next-nearest neighbors of Ni ions. This makes the antiferromagnetic spin structure for the ground state of NiO (https://sundoc.bibliothek.uni-halle.de/diss-online/03/04H009/t3) this cubic structure. NiO is one of the typical systems on which different calculation schemes are tested. It is a charge-transfer insulator with a band gap ~4 eV and local magnetic moment of 1.77μB. This type of magnetic ordering is due to the strong next-nearest-neighbor (nnn) coupling between nickel ions via oxygens 2p shell. The N´eel temperature is NT= 523 K by Barry A. Cipra, Dec 1987.

During experimental studies, difficulties in explaining discrepancies between observations and predictions are encountered due to the fact that high-precision measurements of critical parameters can be unavailable or difficult to obtain.

Some of these difficulties are amenable to solutions using simulation methods. The case of analyzing the properties of phase transition (i.e. the transitions between the paramagnetic and ferromagnetic phase), which is a major problem in experimental studies, is stimulated to make the case.

When using experimental method the following problems must be addressed:

- Experimental studies need special equipment tools in addition to adjusting the experiment to the surrounding environmental conditions

Any experimental result is due to the combined effects of all factors, the strengths and contributions each are practically impossible to single out.

- It is difficult to deal with a complex theoretical statistical mathematical model. Many physical materials undergo phase transitions when they undergo changes in environmental parameters (http://en.mwikipedia.org). Phase transitions of matter are common in physics and familiar in everyday life, the transition of ice to water and water to vapor are familiar examples (World Academy of Science, 2015). Similarly NiO undergoes a phase transition at a specific temperature, that changing its magnetic order from the antiferromagnetic (AF) to the paramagnet state. This magnetic reordering is accompanied by a change in temperature. In other words, the phase transition is a magneto elastic phenomenon (World Academy of Science, 2015).

The square-lattice Ising model is the simplest system showing phase transitions (i.e. the transitions between the paramagnetic and antiferromagnetic (AF) phase). The square lattice Ising model has played a central role in the understanding of phase transitions by Muhammad Wasif and Sabieh Anwar, May 2010.

Phase transition of nickel oxide has been studied using several methods as reported by many researchers. The simulation results have been analyzed and discussed under various deposition conditions as described in many literature reviews. Great efforts have been made by *T. Chatterji et al (2009)* to investigate the antiferromagnetic (AF) phase transition and spin correlations in NiO by high-temperature neutron diffraction below and above NT. They showed that AF phase transition is a continuous second-order transition within their experimental resolution. The spin correlations manifested by the strong diffuse magnetic scattering persist well above NT≈530 K and could still be observed at T=800 K which is about 1.5NT. They argued that the strong spin correlations above NT are due to the topological frustration of the spins on a face-centered cubic (fcc) lattice. The Néel temperature is substantially reduced by this process. They determined that the critical exponents $\beta=0.328\pm0.002$ and $\nu=0.64\pm0.03$ and the N'eel temperature NT=530±1 K. These critical exponents suggested that NiO should be regarded as a 3dXY system. Study of the critical phenomena of Nickel II Iron III oxide (Ferromagnetic) was determined using Monte Carlo simulation technique by *D.A. Ajadi et al (2014)*. The critical temperature (Tc), the magnetization per site (μ), energy per site (E), magnetic susceptibility (χ), specific heat of a $NiOFe_2O_3$ were determined as a function of temperature for two different square lattices 20x20 and 150x150. The analysis of simulation results indicate that the bipolar magnet with strong tetragonal distortion in external magnetic field applied along the axis resembles the behavior of the two dimensional Ising model on the rectangular lattices. The numerical solution of the model in MATLAB "R2013a" was presented. A Monte Carlo Algorithm known as Metropolis Hastings Algorithm was used to evaluate the behavior of the lattice and the critical temperature at which the phase transition between $NiOFe_2O_3$ and paramagnetic state occurs was noted. The analysis of the results shows that TC = 2.25J/KB, in the absence of external magnetic field. It was observed that above (Tc) the material ($NiOFe_2O_3$) becomes a paramagnetic state, and this leads to decreasing in average magnetization and the average Energy increases, while below (Tc) the material is in a ferromagnetic state.

A considerable work has been done by Danny Bennett (2016) by using the Metropolis algorithm. The solutions to various versions of the Ising model were obtained. The 2D square lattice was initially considered. After successfully using the Metropolis algorithm to update the system, the average energy per spin, average magnetization per spin, specific heat capacity and magnetic susceptibility were plotted as functions of temperature in order to gain information about the system. The ground state energy was determined to be -2J, as expected, and the Curie temperature was determined to be TC = 2.6 0.1J/kB in this case, which compares well with the accepted value of TC = 2.269J/kB, and the ground state energy was determined to be -2J. The triangular lattice was then investigated; the ground state energy was determined to be -3J, and the Curie temperature was determined to be TC = 4.2 0.1J/kB. For the 1D system, a phase transition was initially observed, but this was due to a low value of J used in computations; when a larger value of J was used, there was no phase transition, which agrees with the theory. The 3D system was investigated and also determined to be ferromagnetic, with a larger Curie temperature of TC = 4.4 0.1J/kB, and a ground state energy of -3J. Finally, the methods developed in the previous parts of the project were used to investigate a simplified 2D model of NiO. The ground state was determined to be -36:75 0.01meV, and the ordered state of the system was determined to be antiferromagnetic.

In present work, we study the phase transition of Nickel Oxide (NiO) by directly simulate the effect of temperature on its structure. Also to investigate the phase transition of antiferromagnetic phase transition for Nickel Oxide (NiO) using C++ simulation.

2. Theoretical Background

There are some details and concept which will discuss as the following:

2.1 Ising Model

Ising model is a mathematical model of FM in statistical mechanics, which was invented by Wilhelm Lenz (1920). Wilhelm Lenz gave the model as a problem to his student Ernst Ising. Ising solved this problem in one-dimension in his PHD thesis (1924), which worked on linear chains of coupled magnetic moments. Rudolf Peierls named this model by Ising model in his 1936 publication “On Ising’s model of ferromagnetism”. Lars Onsager, winner of the 1968 Nobel Prize in Chemistry, solved two dimensional Ising model in 1944 and exhibited phase transition. And then Ising model enjoyed increased popularity and took its place as the preferred basic theory of all cooperative phenomena by Ajadi et al 2014. A simple classical approximation to an atomic or electronic magnetic moment is provided by an Ising spin which can take two values

$$S_i = \begin{cases} +1 & \text{represents "spin up"} \uparrow \\ -1 & \text{represents "spin down"} \downarrow \end{cases}$$

A two-dimensional magnet can be modeled by a set of N_S spins located on affixed two-dimensional lattice of sites. For example, we can have a square lattice with L spins in the x direction and L in the y direction such $L_2 = N_S$ as shown in figure.1, the up and down arrows represent a positive and negative spin respectively (Xin-Zeng Wu and Di Li Zheng Dai, 2014). The model we will employ in studies of phase transitions (PT) at finite temperature for magnetic systems its simplest form the Hamiltonian is expressed as:

$$H = - J\sum_{<i,j>} S_i S_{j} - B \sum_i S_i \quad (1)$$

With $S_i = \pm 1$, J is a coupling constant expressing the strength of the interaction between neighboring spins and B is an external magnetic field interacting with the magnetic moment set up by the spins. The symbol < ij> indicates that we sum over nearest neighbors only. If the interaction strength J > 0 the system is ferromagnetic: the energy is minimized if the spin point in the same direction Si Sj = +1. If J < 0 the system is antiferromagnetic. B represents an external magnetic field which couples to the magnetization [10]. Periodic boundary conditions are assumed so that each spin $S_{i,j}$ has four nearest neighbors $S_{i+1,j}$, $S_{i-1,j}$, $S_{i,j+1}$ and $S_{i,j-1}$. Each spin interacts with these four nearest neighbors (as shown in figure 2), the dark dot at position (x,y) ,is being interacted upon by its neighbors which are in one lattice spacing from it.) so that it has a potential energy given by

$$E_{i,j} = -JS_{i,j} \{S_{i+1,j} + S_{i-1,j} + S_{i,j+1} + S_{i,j-1}\} \quad (2)$$

The change in energy ΔE due to flip of the spin can simply be (Danny Bennett, Jan2016):

$$\Delta E=2JS_{i,j}\{S_{i+1,j}+S_{i-1,j}+S_{i,j+1}+S_{i,j-1}\} \quad (3)$$

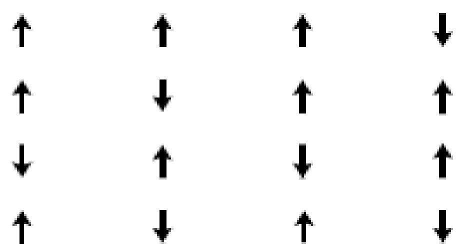


Figure 1. Two Dimensional Lattice of Ising Model

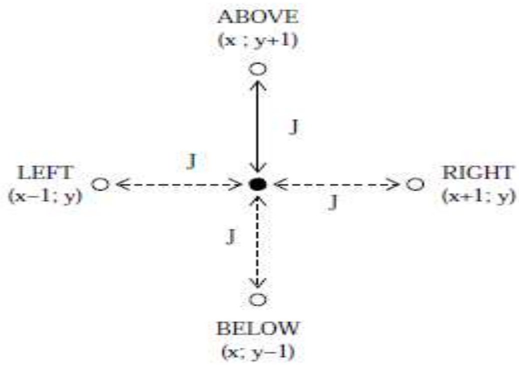


Figure 2.Nearest Neighbor Coupling Spin

2.2 Monte Carlo (MC) Method

Mnote Carlo (MC) simulations involve generating a subset of configurations or samples, chosen using a random algorithm from a configuration space, according to a probability distribution or weight function. Observables are then computed as averages over the samples.

$S_1 = +1$; $S_2 = -1$; $S_3 = +1$.. $S_N = +1$ (4)

In which each spin is set up or down. According to statistical mechanics, the average value of an observable is got by weighting each configuration with the Boltzmann factor. For example, the average magnetization at some fixed temperature T is given by

$$\langle M\rangle = \frac{\sum_{allstates} Me^{-E/K_BT}}{\sum_{allstates} e^{-E/K_BT}} \quad (5)$$

Probability Distribution or weight function is the basic idea of a Monte Carlo (MC) calculation random. The Boltzmann factor is to generate a reasonable number of configurations at exponential function of energy which can vary enormously. The random configurations are therefore generated with probability determined by this exponential factor by Bufallo University, department of physics.

$$P\,(s_1,s_2,\ldots,s_N) = \frac{e^{-E(S_1,S_2 \ldots S_N)/K_BT}}{e^{-E/K_BT}} \quad (6)$$

2.2 Metropolis Algorithm (MA)

The algorithm of choice for solving the Ising model is the approach proposed by *Metropolis et al in 1953*. New configurations are generated from a previous state using a transition probability which depends on the energy difference between the initial and final states. The probability of the nth state for finding the system in a state n is given by

$$P_n(t) = e^{-E_n/K_B\frac{T}{Z}} \quad (7)$$

With energy E_n and Z is a normalization constant which defines the partition function. In the canonical ensemble.

$$Z = e^{-E_n/K_BT} \quad (8)$$

It is difficult to compute since we need all states. In a calculation of the Ising model in two dimensions, the number of configurations is given by 2N with N = L×L the number of spins for a lattice of length L. Fortunately, the Metropolis algorithm considers only ratios between probabilities and we do not need to compute the partition function at all. The algorithm in step form goes as follows:

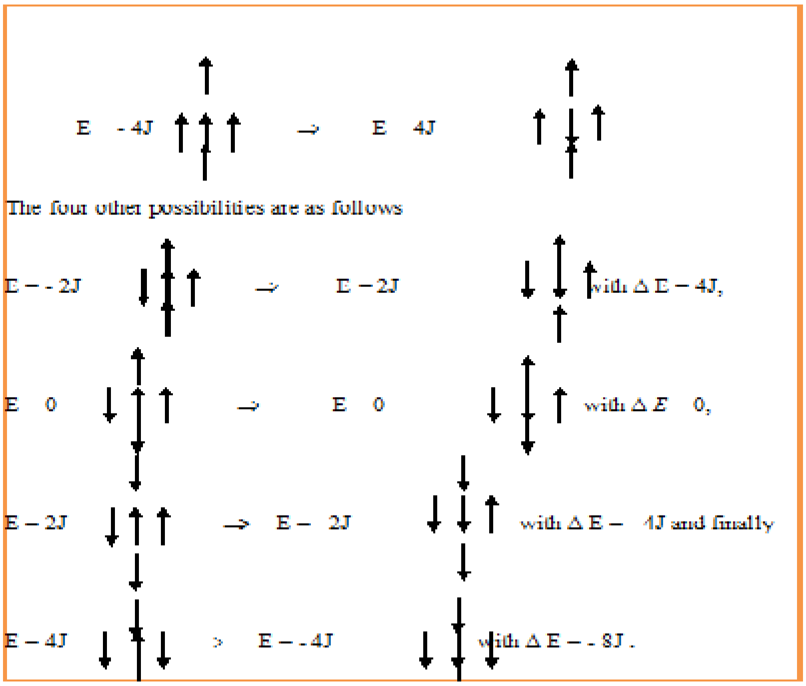
1. Fix the temperature.
2. Establish an initial state with energy E_b by positioning yourself at a random configuration in the lattice.
3. Change the initial configuration by flipping e.g., one spins only. Compute the energy of this trial state E_t.
4. Calculate $\Delta E = E_t – E_b$. The numbers of values ΔE is limited to five for the Ising model in two dimensions see the discussion below.
5. If $\Delta E \leq 0$ we accept the new configuration, meaning that the energy is lowered and we are hopefully moving towards the energy minimum at a given temperature. Go to step 8.
6. If $\Delta E > 0$, calculate $w = e^{(-\Delta E/K_BT)}$
7. Compare w with a random number r ; r in [0,1]. If $r \leq w$

Then accept the new configuration, else we keep the old configuration.

8. The next step is to update various expectations values.
9. The steps (3)-(8) are then repeated in order to obtain a sufficiently good representation of states.
10. Each time you sweep through the lattice, i.e., when you have summed over all spins, constitutes what is called a Monte Carlo cycle. You could think of one such cycle as a measurement. At the end, you should divide the various expectation values with the total number of cycles. You can choose whether you wish to divide by the number of spins or not. If you divide with the number of spins as well, your result for e.g., the energy is now the energy per spin.

The crucial step is the calculation of the energy difference and the change in magnetization. This part needs to be coded in an as efficient as possible way since the change in energy is computed many times. In the calculation of the energy difference from one spin configuration to the other, we will limit the change to the flipping of one spin only. For the Ising model in two dimensions it means that there will only be a limited set of values for ΔE.

Actually, there are only five possible values. To see this, select first a random spin position x, y and assume that this spin and its nearest neighbors are all pointing up. The energy for this configuration is $E = -4J$. Now we flip this spin as shown below. The energy of the new configuration is $E = 4J$, yielding (David P. Landau and Kurt Binder, 2009).



3.3 Curie Temperature (TC)

Curie temperature (TC) is the temperature at which certain magnetic materials undergo a sharp change in their magnetic properties. They lose their characteristic ferromagnetic ability, the Curie temperature for iron is 768°C or 1414 °F. At temperatures below the Curie point the magnetic moments are partially aligned within magnetic domains. As the temperature increase towards the Curie point, the magnetization within each domain decrease. The energy of the atoms are too great for them to join together to form small magnetic areas in the material by Hjorth-Jensen, 2012.

2.4 Phase Transition (PT)

A Phase transition (PT) is basically a natural physical process (*Shenelle Alleyne et al 2009*) it is marked by abrupt macroscopic changes as external parameters are changed, such as an increase of temperature. The point where a phase transition takes place is called a critical point. This section distinguishes normally between two types of phase transitions (PT); first-order transitions and second-order transitions by David P. Landau and Kurt Binder, 2009.

A first order phase transition (PT) is normally accompanied by the absorption or liberation of latent heat, while a second order phase transition has no latent heat. An example of a first order phase transition (PT) is between water and steam (gaseous water), and an example of a second order phase transition that exhibited by is the two dimensional Ising model on a square lattice (magnetization). There is a critical temperature and energy at which the properties of the two phases of the transition are identical. A PT is easily seen on a graph of the spontaneous magnetism versus the temperature of the system as a discontinuity of the data, or a point where the temperature remains constant while the energy input increases (Vincent Anthony Mellor, Jan2011).

3. Simulation Procedure

The two dimensional (2D) square lattice of spins was considered in this work. In the absence of external magnetic field, the Hamiltonian of a spins system is written as:

$$H=-J \sum_{(i,j)}^{N} S_i S_j \quad (9)$$

Where J is the interaction energy of the two spins, N is the total number of spins and Si and Sj are the spin along x and y direction respectively.

The temperature (T) was fixed and then was increased gradually from absolute zero. And at critical temperature Tc, the magnetization changes its state from high value to low value (this is when phase transition occurs).

A Monte Carlo (MC) simulation for 6 x 6 square lattice(SL) of NiO (as represent in figure 3 has been implemented, and from various diagrams the critical temperature (Tc), energy (E), magnetism (M), specific heat (Cv)and magnetic susceptibility (χ) were determined for the square lattice used.

Here it has been assumed that only the nearest neighbors affect each spin (that is, in a two dimension (2D) SL, each spin has four neighbors, up, down, left and right).

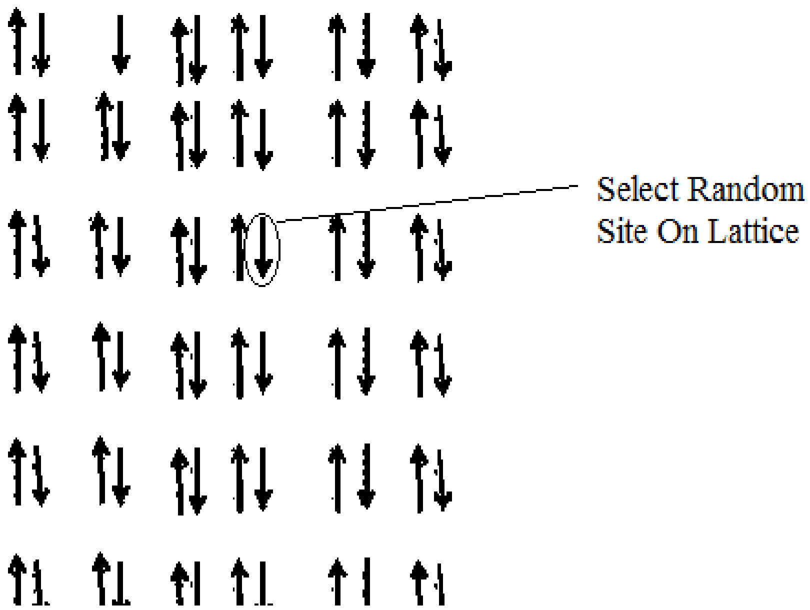


Figure 3. 6X6 Spin Square Lattice

The observables or thermodynamic quantities which were determined in the simulation are energy (E), magnetization (M), energy average ⟨E⟩, average of square energy$\langle E^2\rangle$, magnetization average ⟨M⟩ and average of square magnetization$\langle M^2\rangle$.

The magnetization (M) per spin in a state n was given by

$$M_n = \sum_{i=1}^{N} S_I^n \qquad (10)$$

Since only one spin k flips at a time in the Metropolis algorithm, so the change in magnetization was given by

$$\Delta M = M_m - M_n = \sum_{i=1}^{N} S_I^m - \sum_{i=1}^{N} S_I^n = S_K^m - S_k^n = 2S_k^m;\ S_k^n = -S_k^m \qquad (11)$$

The magnetization average ⟨M⟩ and energy averages were given as follows:

$$< M >= \frac{1}{N_2} \sum_{i}^{N_i} M(S_i) \qquad (12)$$

Select Random Site on Lattice

$$\langle E\rangle = \frac{1}{N_2} \sum_{i}^{N} E(S_i) \qquad (13)$$

Where N_2 is the size of the lattice N_2= 6X6.

Using the Hamiltonian in equation (9) the energy (E) is determined as shown below:

$$E = \sum_{i}^{N_i} H_I = -J\sum_{i}^{N} S_i S_j \qquad (14)$$

$$E_{i,j} = -\{J_1 S_{i,j}\ \{S_{i+1,j} + S_{i-1,j} + S_{i,j+1} + S_{i,j-1}\} + J_2 S_{i,j}\ \{S_{i+1,j} + S_{i-1,j} + S_{i,j+1} + S_{i,j-1}\}\} \qquad (15)$$

Where J_1 and J_2 were the exchange interaction for the NiO and their values are 19.0 meV and -0.4 meV respectively [16].

The change in energy ΔE due to flip of the spin can simply be

$\Delta E = 2J_1 S_{i,j}\{S_{i+1,j} + S_{i-1,j} + S_{i,j+1} + S_{i,j-1}\} + 2J_2 S_{i,j}\{S_{i+1,j} + S_{i-1,j} + S_{i,j+1} + S_{i,j-1}\}$ (16)

For every spin the specific heat (C_V) and the susceptibility (χ) were calculated as follows:

$$C_V = \frac{\partial E}{\partial T} = \frac{< \Delta E >}{K_B T} = \frac{< E^2 >}{K_B T} = \frac{< E^2 > - < E >^2}{K_B T} \quad (17)$$

$$\chi = \frac{\partial M}{\partial T} = \frac{<\Delta M>}{K_B T} = \frac{<\Delta E^2>}{K_B T} = \frac{<M^2> - <M>^2}{K_B T} \quad (18)$$

4. Results and Discussion

This section includes a discussion of results for the effect of temperature on magnetization (M), specific heat C_v and magnetic susceptibility χ of 2D NiO for 6X6 SL.

4.1 Effect of Temperature on Average Energy of NiO

Figure. 4 depicts the average energy was plotted as a function of temperature of NiO square lattice (SL), it is noted that the average energy increases when the temperature increases and from this the ground state was determined to be -38.125 meV. The value of ground state was very close to the result observed by Danny Bennett -36.75 0.01meV [9].

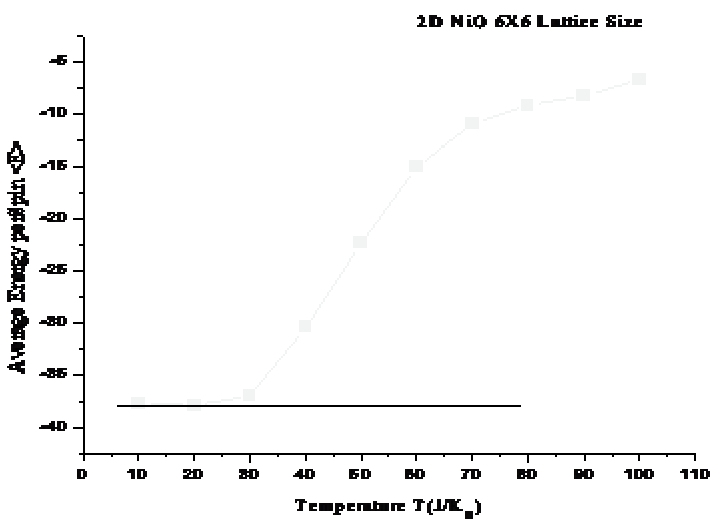


Figure 4. Plot of average energy against temperature T for 6x6 square lattices of NiO

4.2 Effect of Temperature on Average Magnetization of NiO

Figure 5. elucidates the relationship between average magnetization per spin and temperature, it is observed that the magnetization is maximum at low temperatures (ordered state) and vanishes at high temperatures, also the magnetization values decreases from positive to negative value, this points to the fact that the phase transition occurred at the critical temperature Tc≈ 47.2679045 J/KB and the NiO translated from antiferromagnetic to a diamagnetic state.

This result shows a different behavior compared to the results observed by Danny Bennett Jan2016, the difference may be attributed to difference values for exchange interaction and temperature range of NiO (J_1=19 and J_2 = - 0.4, temperature range from 10 to 100 in units of J/KB), while Danny Bennett was took J_1 = 2.3 and J_2 = -21 and low temperature range from 1to 5.

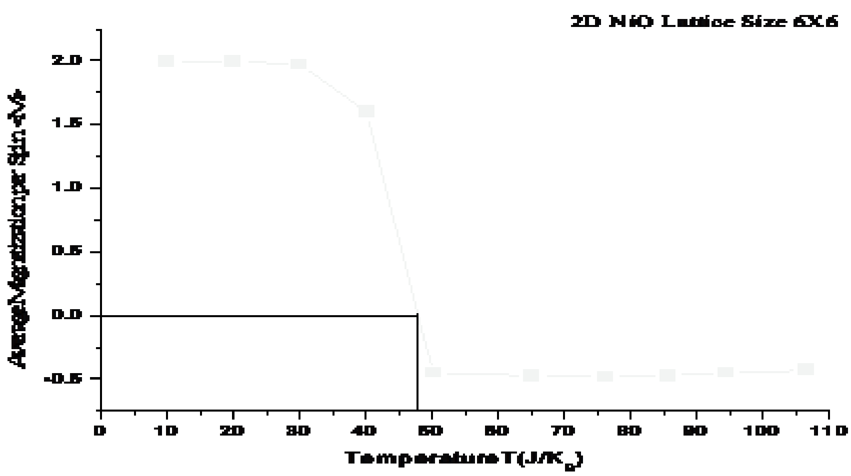


Figure 5.A plot of average magnetization per spin vs temperature of 2D NiO for 6X6 square lattice

4.3 Effect of Temperature on Specific Heat of NiO

From figure 6 it clear that when temperature increases the NiO was transited from antiferromagnetic state to diamagnetic state due to change in specific heat which was observed at critical temperature Tc≈ 47.2679045 J/KB. This is in accordance with what has been observed in the graphs representing the average magnetization is become negative value (see Fig.5).

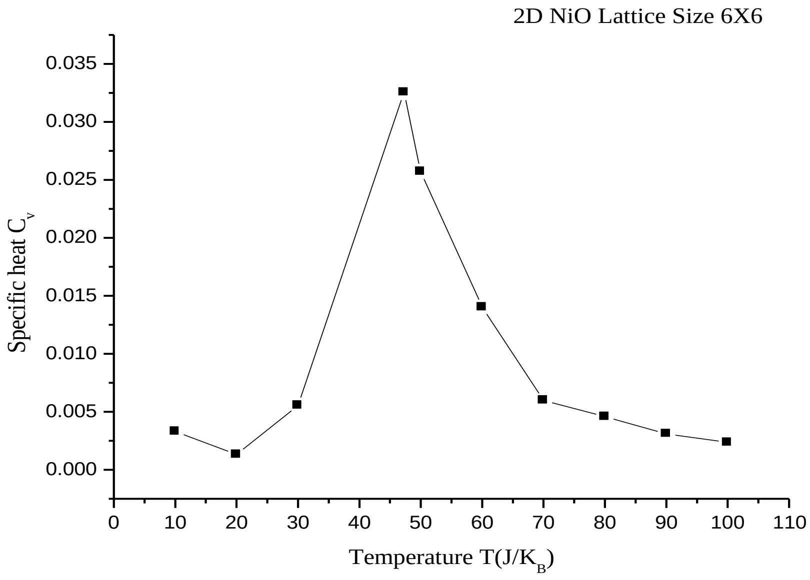


Figure 6 Plot of specific heat Cv VS temperature T for 6x6 square lattices of 2D NiO.

4.4 Effect of Temperature on Magnetic Susceptibility of NiO

Figure 7 illustrates that the magnetic susceptibility is minimum at low and high temperature and has a sharp jump and maximum value at the critical temperature Tc≈ 47.2679045 J/KB. This change for magnetic susceptibility due to change of magnetization (see equation (18)) and also is in accordance with what has been observed in the graph representing the specific heat (see fig 6).

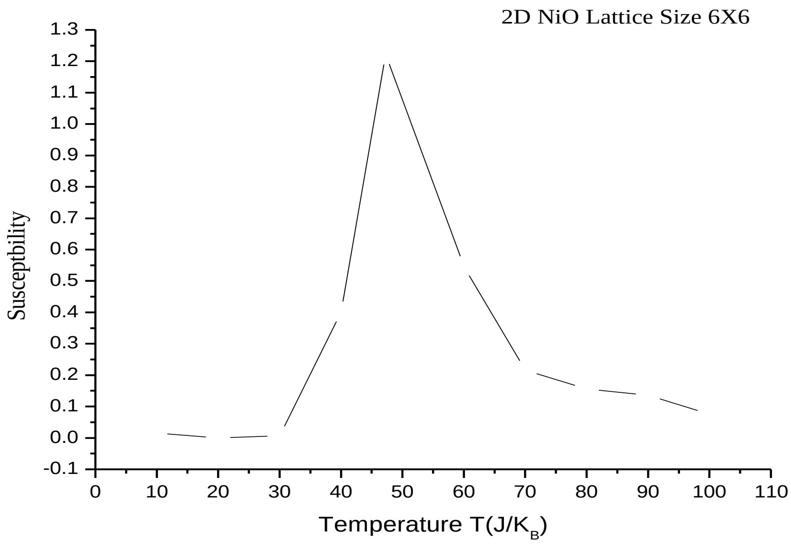


Figure 7. Magnetic susceptibility of a NiO as a function of temperature for 6x6 square lattices.

5. Conclusion

The simulation have started with random spin at the lattice sites and calculated initial magnetization and energy using Ising model. And Metropolis Monte Carlo simulation of an Ising model in C++ was implemented. All the simulations were of 6 x 6 square lattices of NiO, the temperature was run from 10 J/KB to 100 J/KB. After each temperature increment, the system was allowed to equilibrate. The material was described by 2D square lattice having nearest and second nearest neighbor interactions with interaction energies of J_1 = 19 and J_2 = - 0.4, respectively. As the oxygen atoms have no magnetic moment, then it was ignored only the nickel atoms

were considered, and the Boltzmann's constant was set to KB =1. 2D Ising model of NiO was investigated and developed successfully. By plotting the graphs of some thermodynamic functions describing the system such as average energy, average magnetization and specific heat, magnetic susceptibility which were conclude that the system undergoes a phase transition from an antiferromagnetic state to a diamagnetic state at critical temperature Tc≈ 47.2679045 J/KB.

However, the ground state was determined to be -38.125 meV which is very close to the result observed by authors.

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