Monte Carlo Algorithms for Calculating Magnetic Ordering Temperatures

B. R. Billingsley,[1] V. Taufour,[2] and R. Scalettar[2]
[1)] *Physics Department, Washington & Jefferson College*
[2)] *Physics Department, University of California, Davis*

(Dated: 2 January 2021)

Using simulations of the Ising model in three dimensions, we are able to find the magnetic ordering temperature of lattices of varying geometries. We used the Monte Carlo Metropolis algorithm to simulate how the magnet moments of the sites of the Bravais lattices evolve through temperature. With these simulations, we found data of specific heat, Binder ratio, and the magnetic susceptibility as a function of temperature. Our roughly collected data resembled that of precise existing computational data so we can further improve our results by making adjustments to our simulation. This simulation may eventually accelerate the process of finding suitable materials for superconductors and finding the properties of materials may no longer depend so heavily on the time-consuming experimental process.

I. INTRODUCTION

Condensed matter physics encapsulates the study of the physical properties of matter and their electromagnetic interactions on an atomic scale. These studies have paved the way for the formation of new materials that have revolutionized modern technology and civilization such as the discovery of semi-conductors, highfield magnets, and giant magnetoresistant materials, and during the last 200 years, chemists have created new compounds at an exponential 4% growth rate each year.[1]

However, all of these materials' properties are not analyzed at the same rate, and predicting which compounds will result in which properties is difficult to determine. The crystal structure of the material is the first property to be determined, but the determination of other physical properties lags behind. Furthermore, it is hard to predict which materials will be magnetic, and finding the Curie temperature of a magnet without finding it directly through experimentation is not straightforward. If we were able to narrow the search for magnetic materials, we could accelerate the process of finding candidates for superconductors. The most interesting superconductors have been discovered near magnetic instabilities, so finding materials that contain these instabilities may allow us to find superconductors with unconventional properties.

There have been attempts to make these predictions using a relationship between the chemical composition and critical temperature by Nelson and Sanvito. The best of these results produced discrepancies of about 50 K when calculated using machine learning techniques. Nelson and Sanvito grouped around 2,500 known magnets together according to their chemical composition and input the composition data and known Curie temperature data into a machine learning system. This system recognized patterns in the data and was able to predict Curie temperatures for magnets not input in the system. However, they concluded that the best descriptor of a material is the chemical composition so no other factors, such as electronic structure calculations, were applied to potentially improve the predictions.[2]

We now instead attempt to narrow this discrepancy of 50 K by considering the geometry of the compound given by one of the fourteen main Bravais lattices in 3-dimensions shown in

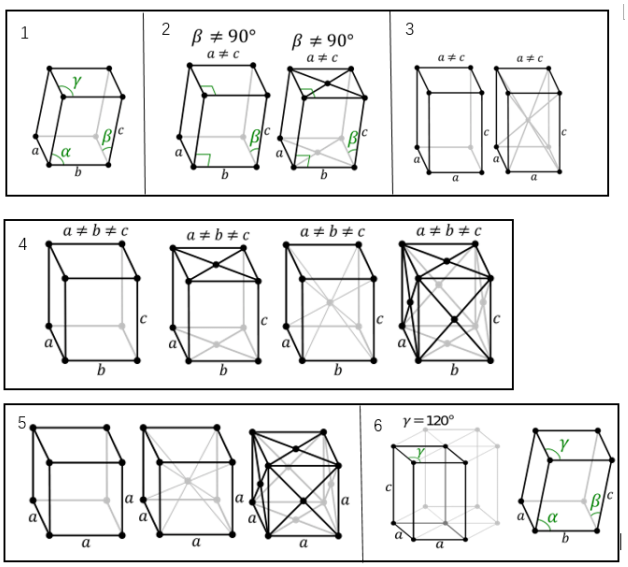

FIG. 1. The fourteen Bravais lattices in 3-dimensions. These geometries are dependent upon the lengths of the sides a, b, and c, and the angles between the edges α, β, and γ. The nearest neighbors are the atoms which are closest in distance to the atom we're looking at, and each of the lattices will have a different set of nearest neighbors. The fourteen lattices are organized into six distinct groups: triclinic, monoclinic, tetragonal, orthorhombic, cubic, and hexagonal.[3]

Fig. 1. This geometrical approach groups materials by configuration and structure as opposed to which elements make up the compound. We can simulate how the magnetic properties of a material of a given geometry will evolve with temperature by analyzing nearest neighbor interactions and how the number of nearest neighbors and the resulting interaction energy varies with geometry. The goal of this research is to produce structural criteria or patterns that dictate magnetic properties through this simulation, and we can simulate this system using the Monte Carlo Metropolis algorithm. This algorithm will allow us to approximate how materials change with temperature as they would in nature by taking into account probabilities and favorable energy configurations.

A. Lattices & Structures of Materials

Materials are composed of a lattice structure of atoms, and this structure can vary depending on the pressure and temperature of the system. These lattices can be unstable or stable, and the properties of the compound depend on this structure. We can determine the lattice structure through different forms of diffraction such as X-ray, neutron or electron diffraction.[4]

The lattice geometry is given by the spacing and angles between the atoms. These lattices have some form of periodicity that is apparent in their configuration as shown in Fig 1. Along with this, the periodicity implies that there is also translational symmetry present along some axis in the lattice. All lattices can be described using a unit cell or a base shape that can be translated to create an entire tessellation.[5] In 2D, this unit cell is defined by two vectors and an angle in between them, and this cell can be applied to every point or atom in the lattice. Bravais lattices are made up of one unit cell repeated for the entirety of the lattice size.

Magnetic materials specifically have their atoms' spins aligned in a distinct way which will vary with temperature. This evolution through temperature can be approximated and described by the Ising model through calculating probabilities and determining the most energetically favorable configuration of spins. Materials at temperatures above their magnetic ordering temperature will have their spins aligned randomly and these materials are not magnetic. When these materials are cooled slowly, they will become magnetized as their spins align in a specific periodic configuration. The material will undergo a phase transition at the magnetic ordering temperature, also called the Curie temperature or critical temperature.

Iron, Nickel, and Cobalt, along with some rare earth elements, are the most magnetic elements on the periodic table. Iron is found in two geometries: face-centered cubic (fcc) and body-centered cubic (bcc). Both of these configurations have a high number of nearest neighbors, fcc having 12 and bcc having 8. Cobalt is found as fcc or hexagonal, both of which have 12 nearest neighbors. Nickel is also in a fcc structure.

B. Ising Model

To simulate the process of finding a material's Curie temperature, we can simulate the Ising Model in 3-dimensions. The Ising Model tells us how a material's magnetic dipole moments (spins) will configure themselves at a given temperature. The model does this by taking into account the probability that a spin will be in a specific configuration due to its interaction with its neighboring spins. The neighboring spins allow the model to determine which configuration is energetically favorable.

At very high temperatures, a material's spins will be randomly aligned and the spins' net alignment will be zero. At high temperatures or when there is large amount of energy in the system, the spins have an equal probability to be be in any configuration or microstate. There are only two configurations for any given spin: spin up or spin down. Due to all of the spins of a lattice having this equal probability, they essentially become randomly aligned.

This probability is related to the partition function, $\mathbb{Z}$. This is given by Eq. (1), and is a sum of the Boltzmann factor at a given energy level over all possible energy levels. In Eq. (1), E_i is the energy at a given state i, β is $\frac{1}{k_B T}$ where k_B is the Boltzmann constant, and T is the temperature.

$$\mathbb{Z} = \sum_i e^{-\beta E_i} \tag{1}$$

With the partition function, we can find the average energy, and the average energy is dependent upon the probability of the atom or particle to be in that energy level. The probability is given by the Boltzmann factor divided by the partition function. This relationship is shown in Eq. 2 where $P(E_i)$ is the probability of the atom to be in that energy state.

$$\begin{aligned} \langle E \rangle &= \sum_i E_i P(E_i) \\ \langle E \rangle &= \frac{\sum_i E_i e^{-\beta E_i}}{\mathbb{Z}} \end{aligned} \tag{2}$$

As we decrease temperature, less energy is present in the system so not all of the microstates will have the same equal probability. This unequal probability leads the spins to want to align in an energetically favorable configuration which takes into account the spins of its nearest neighbors. If the neighbors are all spin up, it is energetically favorable to align in the spin up direction.

In the case of magnets, spins will align in the same direction or a form of periodic arrangement. For example, ferromagnetic materials have all of their spins in the same direction, paramagnetic materials will have an induced magnetization in the presence of a magnetic field, and anti-ferromagnetic materials will have a periodic pattern of spin up and spin down. The nearest neighbor interaction is also defined by the interaction energy, J. This describes the energy between two neighbors at a certain distance so J for all of the nearest neighbors will be the same value. In the case of ferromagnets, $J > 0$, and in anti-ferromagnets, $J < 0$.

The temperature where there is a net magnetization or a majority of the spins become aligned and undergo the phase transition is the magnetic ordering temperature. We can find the Curie temperature experimentally through measuring the specific heat, the intersection of the Binder ratio of different size lattices, or the magnetic susceptibility, χ. The Binder ratio is a dimensionless ratio of average magnetization which becomes size independent at the Curie temperature. This means that for any size lattice, we find that the value of the Binder ratio is the same at the Curie temperature. For our simulations, this is the most reliable way to find the Curie temperature because of its lack of dimensions and exact finding of the Curie temperature. The specific heat and magnetic susceptibility data will give us a rough estimate of where the phase transition occurs, but it cannot give us an exact value like the Binder ratio can.

The specific heat, C, is given by Eq. 3 where E is the energy of the system. The Binder ratio, B, is given by Eq. 4 where M

is the magnetization of the system, and magnetic susceptibility, χ, is given by Eq. 5.

$$C = \frac{\langle E^2 \rangle - \langle E \rangle^2}{T^2} \tag{3}$$

$$B = \frac{3}{2} - \frac{\langle M^4 \rangle}{2\langle M^2 \rangle^2} \tag{4}$$

$$\chi = \frac{\langle M^2 \rangle - \langle M \rangle^2}{T} \tag{5}$$

II. COMPUTATIONAL METHODS

To be able to create this Ising model simulation, we primarily used Python and FORTRAN, and we used the Monte Carlo Metropolis algorithm to simulate how each spin would evolve through temperature.

We can use the enumeration and partition function method for small lattices as we can quickly find all of the microstates. We can create either a chain or 2-dimensional lattice of up and down spins represented by 1 and -1 respectively and then find all the microstates of this system. The microstates are the number of combinations of 1 and -1 we can have. The number or microstates is given by 2^N where N is the number of spins. We could find the partition function and then for each microstate, calculate and plot the average energy, specific heat, and entropy as a function of temperature.

This enumeration technique has obvious limitations depending on lattice size. A 4x4 lattice of 16 spins will have 2^{16} or over 65,000 microstates. The time a computer takes to perform these operations can be given by the multiplication of the number of microstates and the number of operations required to compute E, which is about 100, then divided by the computer processing speed. In this case, it would be $(65,000)(100)/(3\times10^9)$. A household computer can actually compute a 4x4 lattice in less than a second, but for an 8x8 lattice with 64 spins, this would take 20,000 years to compute.

Given this, we become dependent on another method to compute regular size lattices, and we can use Monte Carlo techniques to simulate our lattice instead.

A. Monte Carlo Metropolis Algorithm

Monte Carlo algorithms can be used to measure physical quantities as the output from these algorithms are similar to those results found in nature. Most importantly, the algorithm can find these quantities without finding a partition function.[6]

The algorithm utilizes random number generation and probability to determine whether or not changes to the system are accepted or rejected. The Metropolis algorithm was created for systems with two or more interacting atoms.[7] Our implementation of the algorithm assumes periodic boundary conditions on a finite lattice of N particles. Creating a lattice with $N = 10^6$ for example is easy to do, and we can then extrapolate $N \to \infty$ using the Binder ratio.

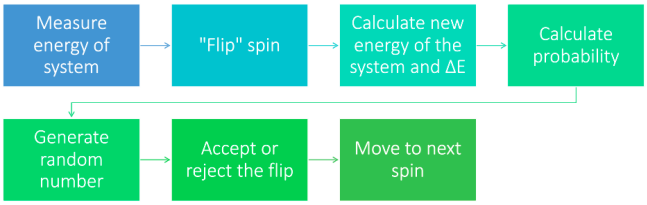

FIG. 2. A visualization of the process the Metropolis algorithm goes through to either accept or reject a change to the system.

Specifically, our simulation creates a random 2D lattice of size nxm containing elements of -1 and +1. Our simulation assumes that $\beta = 1$ and $J = 1$. The initial energy of the system is calculated using Eq. 6 where J_{ij} is the interaction energy between two spins, i and j. Variable s_i is the value of the first spin and s_j is the value of the neighbor being evaluated. So s_i and s_j will only ever be ±1. Since we are only looking at nearest neighbors, the interaction energy for all nearest neighbors will be the same, so we can factor this out of the summation to result in Eq. 7. The total energy of the lattice is given by this sum over every spin in the system.

$$E = -\sum_{\langle i,j \rangle}^{N} J_{ij} s_i s_j \tag{6}$$

$$E = -J\sum_{\langle i,j \rangle}^{N} s_i s_j \tag{7}$$

The algorithm begins by looking at one spin and flipping the spin to opposite its original value. It then calculates the change in energy, ΔE, by comparing the difference in energy before and after the flip. With this change in energy, a probability is calculated using Eq. 8. This probability tells us if this change is energetically favorable. This is not a true probability because this value can be greater than 1, but it will only be greater than 1 when the overall energy of the system is decreased by the flip.

$$P = e^{-\Delta E/T} \tag{8}$$

At this time, the simulation generates a random number between 0 and 1. If the random number is less than our probability, then the flip is accepted, and if the random number is greater than the probability, the flip is rejected and reverted back to its original configuration. Since our probability can be greater than 1 in the case of a lower energy state, these flips are always accepted. Once the flip is either accepted or rejected, we move on to the next spin in the lattice and repeat this process. This can also be shown in the flowchart in Fig. 2. After sufficient sweeps of going through the lattice that was originally randomized, we find that the spins align as temperature decreases and we see groups of spin up and spin down which resemble ferromagnetic domains!

It is important to note that even though a site may be surrounded by spins of the same alignment, and the energetically

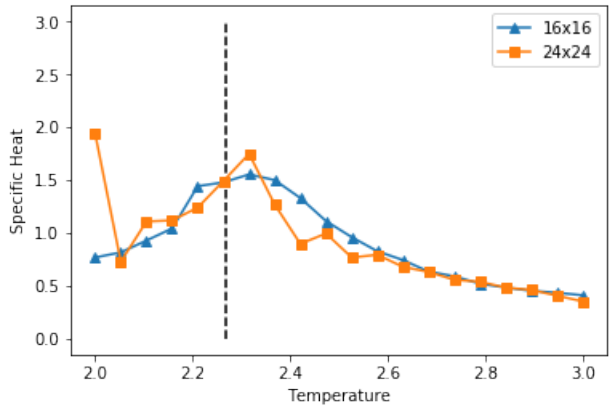

FIG. 3. Ten thousand sweeps of a 16x16 and 24x24 size lattice using the Monte Carlo simulation. The dotted line represents the known Curie temperature for comparison to the peak value of the curve. It can be seen that the 24x24 lattice is noisy and it would be difficult to extract the peak of the curve with only this data. This data took an hour and thirty minutes to collect in Python.

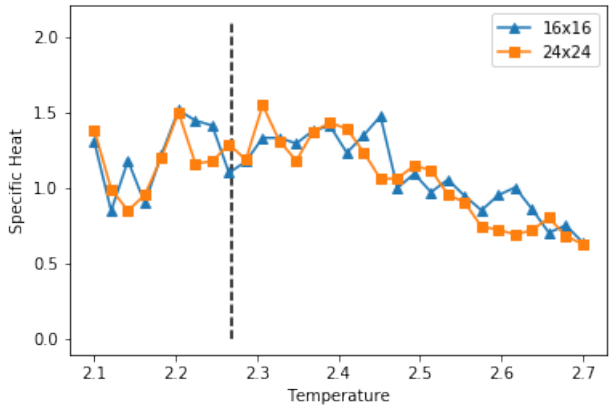

FIG. 4. One thousand sweeps of a 16x16 and 24x24 size lattice using the Monte Carlo simulation. It can be seen that while this data has the expected shape, there was an insufficient number of sweeps through the lattice so the data is too noisy to be able to be used or analyzed.This data took ten minutes to collect in Python.

favorable position would be to stay aligned, there is still a small probability that the flip will be accepted in the opposite direction.

Every time the simulation goes through all points in the lattice, this is one sweep. When moving onto the next sweep, we use the final lattice of the previous sweep as the original lattice of the next sweep. At the end of a given number of sweeps, we decrease the temperature and repeat using the final lattice once more. With each temperature, we measure the energy and magnetization and keep track of how these values change through temperature and flips. The energy is kept as a running update of the original energy; we subtract the change in energy if the flip is accepted or it stays the same if rejected. We measure magnetization by summing up all of the spins in the lattice.

When we run this simulation, we find the same results from that of the enumeration technique. Because of this, the number of operations and iterations of the program is lessened, and we can do much larger lattices. However, if we do not perform a sufficient number of sweeps through the lattice, our data will be noisy and inaccurate. An example of this is shown in Fig. 3 and Fig. 4. It can be seen that with only 1,000 sweeps through the lattice, the proper shape is present, but without context, it would be difficult to determine where the peak is.

B. Efficiency in Python

Even with 10,000 sweeps of a lattice, we would ideally need 100,000 sweeps for most accurate data. However, although Monte Carlo is more efficient than enumeration, we find that the coding language Python is not equipped well enough to be able to handle this number of sweeps.

We found that determining the neighbors of each site as we go through the lattice is time consuming. Our program had referred back to a function to find the four surrounding neighbors in the case of a square lattice, and the act of calling back to this function, regardless of how complicated the function is, makes the code run that much more slowly. Similarly, we found that calculating the random numbers as we need them is inefficient. For both of these issues, we find that pre-calculating the neighbors and random numbers eliminates this issue. So we create a neighbor table for all directions in a lattice that contain the location of say the north neighbors in a list. We also would have a list of all of the random numbers.

We also found that we actually don't need to sweep through every single lattice point to be able to simulate our lattice. In the original Metropolis paper, they note that we should instead perform a random sampling of points in the lattice. This gives us the same result as if we had gone through every point.

Despite these changes, we ultimately transitioned our codes to FORTRAN, a computer language that can perform the same computations much more efficiently without these numbers of workarounds.

III. RESULTS & INTERPRETATION

When finding the Curie temperature, T_C, we first want to ensure that our simulation works by comparing our output to known data. To do this, we ran a simulation of four different size lattices of the simple cubic in 8^3, 12^3, 16^3, and 20^3. This was run roughly to see if we would get the desired shape and to acknowledge any errors or bugs in the code before moving onto the next Bravais lattice with an unknown T_C. The simple cubic lattice has 6 nearest neighbors and a known T_C of 4.51 K·J. This isn't in terms of temperature, but instead a fraction dependent on the interaction energy. Our results for specific heat, Binder ratio and magnetic susceptibility can be seen in Fig. 5, Fig. 6, and Fig. 7. The number of nearest neighbors or coordination number of some of the Bravais lattices is shown in Table I.

We can see that this data, although rough and imprecise,

Bravais Lattice	# of Nearest Neighbors
Simple Cubic	6
Face-Centered Cubic	12
Body-Centered Cubic	8
Triclinic	2
Primary Monoclinic	2
Base-Centered Monoclinic	4
Hexagonal	12

TABLE I. The number of nearest neighbors and the corresponding lattice. Not all of the data has been collected yet, and neither have the number of nearest neighbors. It can be seen that both triclinic and primary monoclinic have 2 nearest neighbors, so there is no distinguishing factor between the two lattices in the simulation as it stands.

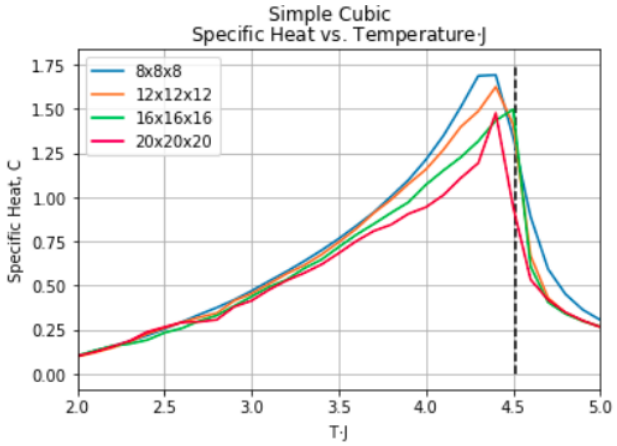

FIG. 5. The specific heat as a function of temperature of a simple cubic lattice with a coordination number of 6. The dashed line represents the known value for T_C. The specific heat should peak around T_C, but not peak on the exact value, so this result matches well. We also see that as the lattice size increases, the peaks approach T_C.

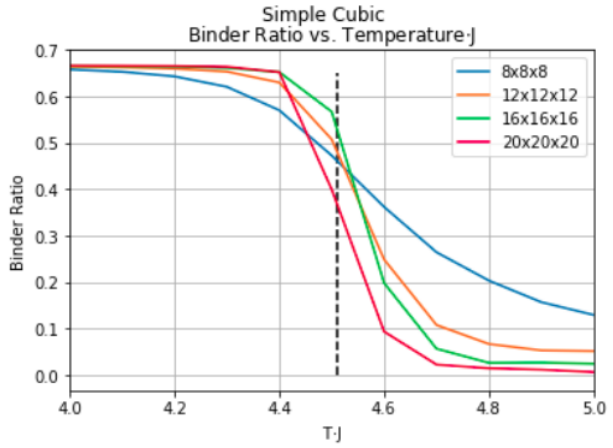

FIG. 6. The Binder ratio as a function of temperature of a simple cubic lattice with a coordination number of 6. The dashed line represents the known value for T_C. Since Binder ratio is size independent at T_C, we should find an intersection of different size lattices at this point. We see a crossing around the dashed line which we can assume would have matched well with a more precise run of data with a smaller change in temperature.

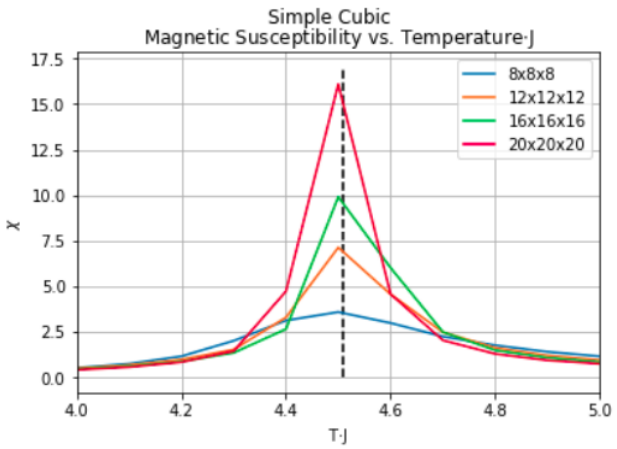

FIG. 7. The magnetic susceptibility as a function of temperature of a simple cubic lattice with a coordination number of 6. The dashed line represents the known value for T_C. The magnetic susceptibility should peak around T_C which this data does.

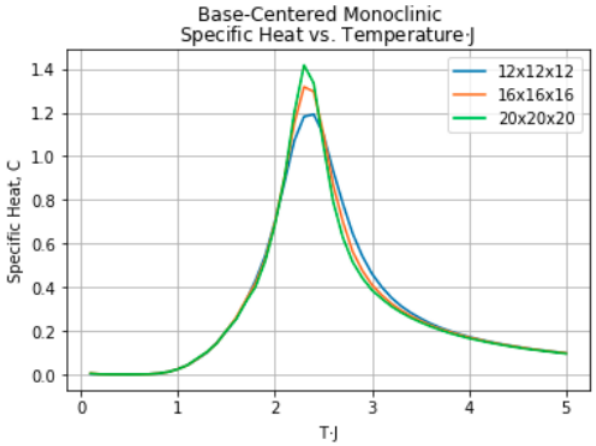

FIG. 8. The specific heat as a function of temperature of a base-centered monoclinic lattice with a coordination number of 2. The specific heat should peak around T_C, so we can see that T_C may be between 2 and 3 K·J.

does match the theoretical value. When measuring the specific heat and magnetic susceptibility, we expect a peak around the Curie temperature, so this data is good for showing the approximate range where T_C occurs. However, because the Binder ratio is size independent at T_C, a very precise run of several sized lattices with the same parameters should produce a graph showing an intersection at the Curie temperature. This is our most exact way of determining T_C.

Knowing that the simple cubic simulation produced values we expect, we can be sure that our code works and can be applied to the other lattices. When running the simulation for the base-centered monoclinic, we result in the data shown in Fig. 8, Fig. 9, and Fig. 10.

For the base-centered monoclinic it can be seen from the data that the Curie temperature occurs around 2 K·J. To find the exact value, we will need to run more sweeps of the lattice

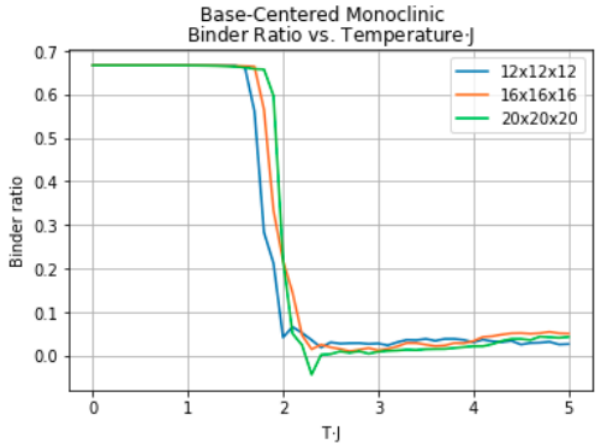

FIG. 9. The Binder ratio as a function of temperature of a base-centered monoclinic lattice with a coordination number of 2. We see a crossing between 1.75 and 2.25 K·J.

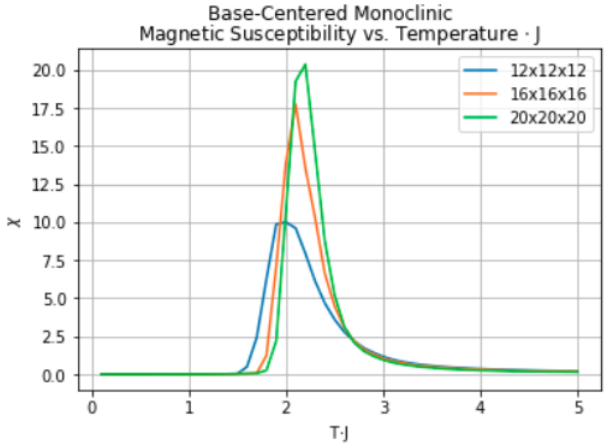

FIG. 10. The magnetic susceptibility as a function of temperature of a base-centered monoclinic lattice with a coordination number of 2. The magnetic susceptibility should peak around T_C, so this shows us that T_C may be between 1.75 and 2.5 K·J.

in the simulation. From this data, though, we are able to see trends and now we have a rough idea of where the Curie temperature is, so there is less guessing involved when we refine the simulation.

For both sets of data, more precise measurement is necessary. These were taken with 100,000 sweeps at a temperature increment, dT, of 0.1 K. This causes us to have large gaps at important data ranges like that of the Curie temperature. When rerunning this simulation, we will want to take more data points and for more sweeps at each temperature.

Similar graphs can be created for for the other 12 Bravais lattices. The data produced from the primary monoclinic simulation shows a Curie temperature around 1 K·J. While this isn't anything more than an estimate at this stage, it is interesting to note that lattices with more neighbors are showing to have a higher Curie temperature than those with fewer neighbors. This is a promising trend if we want to correlate the coordination number to the properties of the material.

IV. CONCLUSIONS

The only parameter that changes in the simulation when run for different Bravais lattices is the nearest neighbors that are input. We input a neighbor table that contains all of the directional information for the array to be able to select the neighbors in the energy calculations. The simulation will output the data of our physical quantities we are trying to find. An advantage and limitation of our simulation is that our program is only dependent on the number of neighbors a site has. So, as our simulation is currently written, regardless of which neighbors a site has, that lattice will always produce the same result as another lattice with the same coordination number. To be able to distinguish between two Bravais lattices with the same coordination number, we need to add secondary neighbors that will have a different corresponding interaction energy.

Additionally, to advance our simulation, we can add uniaxial pressure variation to the system and vary the interaction energy with this to see how the properties change. Adding pressure may increase the interaction energy since the spins would then be closer together.

Although this work is still in progress, we are able to see that our simulation of the Ising model using the Monte Carlo Metropolis algorithm produces results to find the Curie temperature of the different Bravais lattices. With future work refining the simulation to get more precise data, we will be able to find the Curie temperature with less of a range of error.

ACKNOWLEDGMENTS

We wish to acknowledge the NSF's Research Experience for Undergraduates program at the University of California, Davis for providing funding for this research.

REFERENCES

[1] E. J. Llanos, W. Leal, D. H. Luu, J. Jost, P. F. Stadler, and G. Restrepo, "Exploration of the chemical space and its three historical regimes," **116**, 12660–12665 (2019).
[2] J. Nelson and S. Sanvito, "Predicting the curie temperature of ferromagnets using machine learning," Phys. Rev. Materials **3**, 104405 (2019).
[3] T. Zhang, L. Li, and H. Yang, "3*d* crystal image analysis based on fast synchrosqueezed transforms," Awaiting Publication (2018).
[4] D. C. Palmer, "Crystal structure determination, 2nd ed." Geological Magazine **142**, 306–307 (2005).
[5] A. S. Phani and M. Hussein, "Introduction to lattice materials," (2017) pp. 1–17.
[6] G. S. Beddard, "Using the metropolis algorithm to calculate thermodynamic quantities: An undergraduate computational experiment," Journal of Chemical Education **88**, 574–580 (2011).
[7] N. Metropolis, A. W. Rosenbluth, M. N. Rosenbluth, A. H. Teller, and E. Teller, "Equation of state calculations by fast computing machines," The Journal of Chemical Physics **21**, 1087–1092 (1953), https://doi.org/10.1063/1.1699114.