THERMODYNAMIC PHASE TRANSITION OF A MAGNETIC SYSTEM Curie Temperature Predicted by the Monte Carlo Method

by

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Original scientific paper
https://doi.org/10.2298/TSCI2004295D

Curie temperature is an important parameter in the second-order thermodynamic phase transition of a magnetic system. However, the classical Heisenberg's mean field theory tends to overestimate heavily the temperature. In order to solve this problem, firstly, the structure of ferromagnetic and spin-glassy materials in a magnetic system is established by the Ising model. Secondly, the respective energy of ferromagnetic and spin glass states is calculated by Monte Carlo method. Finally, Curie temperature is predicted through the obtained energy, which agrees well with experimental data. A new strategy to estimate accurately Curie temperature is presented.

Key words: *Monte Carlo method, thermodynamics, phase transition temperature, needle throwing experiment*

Introduction

Magnetic materials will undertake a harsh change in their magnetic properties at Curie temperature, T_c, which belongs to the second-order thermodynamic phase transition and has an important physical significance. In the 19th century, statistical physicists began to study T_c, but the work did not attract much attention. Until 1996, Ohno *et al.* [1] obtained a diluted magnetic semiconductor (DMS) $Ga_{0.95}Mn_{0.05}As$ film with T_c as high as 110 K by the low temperature molecular beam epitaxy (LT-MBE), the DMS combines magnetic and semiconductor properties, causing great interest among researchers. Experimentally, all the reported semiconductors with ferromagnetism are *p*-type semiconductors [2-5], unfortunately, which persist ferromagnetism at a low temperature. For example, the highest T_c of DMS GaMnAs is 150 K [6], while T_c of GaMnN ranges from 228 to 370 K [7]. In tradition, DMS with high T_c is fundamental to build practical spintronic devices that could work at room temperature. Therefore, the theoretical prediction of T_c has emerged. Dietl *et al.* [8] predicted T_c of 13 semiconductors doped with Mn by the Zener model, resulting in $T_c \sim 120$ K for $Ga_{0.95}Mn_{0.05}As$. Meanwhile, GaAs doped 5% Mn shows a T_c near 260 K calculated by Heisenberg mean field theory [9]. Recently, the response of external parameters (such as strain and pressure) on magnetic properties was reported [10-12]. Furthermore, 2-D ferromagnetic crystal (FM) with high T_c was also investigated [13]. Heisenberg model [9], Ising model [13, 14], and Monte Carlo model [15] were often used. The geometric potential theory [16-21] can explain the effect of FM ordering in FM materials on its FM property [19].

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In this work, Ising model combined with Monte Carlo algorithm is proposed to improve the algorithm of the Heisenberg model, the obtained T_c value is closer to the experimental one. Finally, we explain why the T_c value estimated in [9] is much higher than the corresponding experimental result.

Thermodynamic theory of Curie temperature

The T_c is the second-order thermodynamic phase transition temperature of magnetic materials. When the material temperature is below T_c, the material shows ferromagnetism, whereas, the material exhibits paramagnetism. In statistical mechanics, the theory of entropy change is used to study T_c, but it is not applicable to the prediction of T_c of practical materials, so the Zener model, Heisenberg model and Ising model were developed to predict T_c.

Heisenberg model

According to the Heisenberg model [9], T_c can be expressed:

$$K_B T_c = \frac{2}{3}\frac{\Delta E}{x} \tag{1}$$

where K_B is the Boltzmann constant, x – the doping concentration, and ΔE – the difference between the bonding energy of spin glass state and the bonding energy of the FM state.

The key to calculate T_c by the Heisenberg model is to obtain the bonding energy of the FM state and spin glass state. The bonding energy is mainly based on first-principle calculations. The bonding energy of the FM state can be easily obtained. However, there are some problems in calculating the bonding energy of spin glass states, for example:

– The first-principle calculations are used to model the spin glass state, which can only be described by antiferromagnetic (AFM) state, thus, the resulting T_c is much higher than that of experimental results. For instance, GaAs doped with 5% Mn is estimated to have the value of T_c as high as 260 K [9], whereas only 110 K was obtained experimentally [2]. Recently Wu *et al.* [22] also fabricated a material with a Curie temperature of 237 °C.
– The spin glass state is described as half of the spin direction of magnetic ions is upward and the other half is downward. However, the spin direction of magnetic ions is half up and half down, which can be simulated in many ways. For example, in [15], three AFM states are used to calculate the bonding energy of the system, so there are three T_c calculations, we have no criterion to judge which one is right or all three values are incorrect.
– The spin glass state is disordered, the spin direction is random, and the AFM state is orderly, so it is not advisable to use an AFM state to describe the spin glass state. The first-principle calculations cannot realize the random distribution of spin direction, which is the limitation of the Heisenberg model in applications.

Ising model

As for the origin of magnetism, according to the conclusion of [2, 3], magnetism is caused by holes, and the following models are established: Let the system have N cells, n holes after doping, then $x = n/N$ is the doping concentration. Each hole is a magnetic moment, then there are n magnetic moments, the bonding energy of the system can be calculated by the interaction of magnetic moments. In this work, the bonding energy is obtained by using the Ising model [13, 14] under the condition that only the nearest neighbor interaction is considered.

In the Ising model [13, 14], see fig. 1, each site i, $i = 1,\ldots,n$, has a spin S_i which can take on the values ±1. Nearest-neighbor sites contribute an energy $-JS_iS_{i+1}$, where J is the exchange parameter, then Hamiltonian is:

$$H = -\frac{1}{N} J \sum_{i=1}^{n} S_i S_{i+1} \tag{2}$$

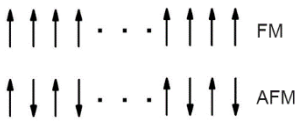

Figure 1. The FM and AFM states

From this, the bonding energy of FM state and AFM state can be calculated.

Calculation of bonding energy of three states

Calculation of bonding energy of FM state and AFM state by Ising model

According to eq. (1), when using the Heisenberg model and first-principle calculations to calculate bonding energy, AFM state is used instead of spin glass state, which will inevitably lead to errors. In order to reduce the error, here we adopt the Monte Carlo algorithm to calculate the bonding energy of the spin glass state based on the Ising model. From eq. (2), the bonding energy of FM state and AFM state can be obtained:

$$E_{\mathrm{FM}} = -\frac{1}{N} J \sum_{i=1}^{n} S_i S_{i+1} = -Jx \tag{3}$$

$$E_{\mathrm{AFM}} = \frac{1}{N} J \sum_{i=1}^{n} S_i S_{i+1} = Jx \tag{4}$$

Then, the energy difference ΔE_1 and T_{c} are:

$$\Delta E_1 = E_{\mathrm{AFM}} - E_{\mathrm{FM}} = 2Jx \tag{5}$$

$$T_{\mathrm{c}_1} = \frac{2}{3} \frac{\Delta E_1}{x K_{\mathrm{B}}} = \frac{2}{3} \frac{2J}{K_{\mathrm{B}}} \tag{6}$$

Calculation of bonding energy of spin glass state by Monte Carlo algorithm

The key to calculating the bonding energy of the spin glass state by Monte Carlo algorithm is to generate high quality random numbers, and to apply the generated random numbers to the spin glass state model. In order to solve these two problems, this paper uses MATLAB to generate random number "1" or "–1", where "1" means spin up, and "–1" means spin down. According to the sequence of random number generation, S_i in the Ising model is assigned in order. From this, spin glass states with random distribution can be generated, and the bonding energy can be calculated.

According to Monte Carlo algorithm, MATLAB program is compiled to calculate the bonding energy of spin glass state is that $E_{\text{spin glass}} = 0$, then the energy difference and T_{c} are:

$$\Delta E_2 = E_{\text{spin glass}} - E_{\mathrm{FM}} = Jx \tag{7}$$

$$T_{c_2} = \frac{2}{3}\frac{\Delta E_2}{xK_B} = \frac{2}{3}\frac{J}{K_B} \tag{8}$$

Form eqs. (5)-(8), we get:

$$\begin{cases} \Delta E_1 = 2\Delta E_2 \\ T_{c_1} = 2T_{c_2} \end{cases} \tag{9}$$

It can be seen that T_c calculated by using AFM bonding energy is twice as high as that calculated by using spin-glass bonding energy. This leads to the fact that the T_c calculated by first-principle calculations is always much higher than the one of experiment.

For example, in [9] estimated a value of T_c about 260 K for $Ga_{0.95}Mn_{0.05}As$ by first-principle calculations, while FM only persists up to 110 K [2]. There exists a big gap between the theoretical and the experimental results. Conversion of T_c using eq. (9) yields $T_{c_2} = T_{c_1}/2 = 130$ K, which is closer to the experimental value of 110 K.

Needle throwing experiment to verify the bonding energy of spin glass state

During calculation of the bonding energy of spin glass state in eq. (2), the spin glass state is simulated by using the random number sequence "1" and "–1" generated by MATLAB programming, and the bonding energy calculated is 0. In this section, the classical needle throwing experiment is used to further confirm that the bonding energy of spin glass state is 0.

As shown in fig. 2, we drew parallel lines of 1 cm on a piece of white paper with 50 × 50 cm, which was then placed on a horizontal table. A pin directing the *z*-axis was fallen freely at a height of 1 m, the angle between the pin point and the *x*-axis was measured. If the angle is less than 90°, the spin direction is upward, and the spin value is assigned according to the Ising model, if the angle is greater than 90°, the spin direction is downward, and the spin value is assigned according to the Ising model, if it is perpendicular to the *x*-axis, this data is discarded. In this paper, a total of 2000 needles were thrown and 2000 data were available. The data were used to simulate the spin glass state. The calculated bonding energy was 0.004, which could be approximately equal to 0. Thus, this experiment was a success, and the correctness of Monte Carlo algorithm in eq. (2) is confirmed again by the needle throwing experiment.

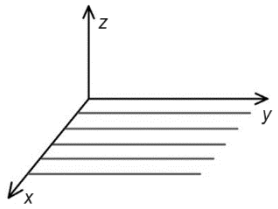

Figure 2. Sketch map of needle throwing experiment

Conclusion

In this paper, based on the Ising model and the Monte Carlo algorithm, we proved that the energy difference between the FM state and the AFM state is twice of that between the FM state and the spin glass state. The problem of substituting AFM state for spin glass state in bonding energy calculation based on the Heisenberg model and first-principle calculations is solved. This work suggests an improved algorithm for accurate T_c calculation.

Acknowledgment

The research work is supported by the National Natural Science Foundation of China (11861049) and the Natural Science Foundation of Inner Mongolia (2017MS0101, 2018MS01027).

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Paper submitted: December 26, 2018
Paper revised: June 19, 2019
Paper accepted: August 18, 2019