The Study of the Influence of Matrix, Size, Rotation Angle, and Magnetic Field on the Isothermal Entropy, and the Néel Phase Transition Temperature of $\text{Fe}_2\text{O}_3$ Nanocomposite Thin Films by the Monte-Carlo Simulation Method

Dung Nguyen Trong $^1$, Van Cao Long $^1$ and Ștefan Țalu $^2$

1 Institute of Physics, University of Zielona Góra, Prof. Szafrana 4a, 65-516 Zielona Góra, Poland; vancaolong2020@gmail.com
2 The Directorate of Research, Development and Innovation Management (DMCDI), Technical University of Cluj-Napoca, 15 Constantin Daicoviciu St., 40020 Cluj-Napoca, Romania; stefan_ta@yahoo.com
* Correspondence: dungntdt2018@gmail.com

Abstract: In this paper, the study of the influence of the matrix structure (mxm) of thin-film, rotation angle ($\alpha$), magnetic field (B), and size (D) of $\text{Fe}_2\text{O}_3$ nanoparticle on the magnetic characteristic quantities such as the magnetization oriented z-direction ($M_{zE}$), z-axis magnetization ($M_z$), total magnetization ($M_{\text{tot}}$), and total entropy ($S_{\text{tot}}$) of $\text{Fe}_2\text{O}_3$ nanocomposites by Monte-Carlo (MC) simulation method are studied. The applied MC Metropolis code achieves stability very quickly, so that after 30 Monte Carlo steps (MCs), the change of obtained results is negligible, but for certainty, 84 MCs have been performed. The obtained results show that when the mxm and $\alpha$ increase, the magnetic phase transition appears with a very small increase in temperature Néel ($T_{\text{Ntot}}$). When B and D increase, $T_{\text{Ntot}}$ increases very strongly. The results also show that in $\text{Fe}_2\text{O}_3$ thin films, $T_{\text{Ntot}}$ is always smaller than with $\text{Fe}_3\text{O}_4$ nano and $\text{Fe}_2\text{O}_3$ bulk. When the nanoparticle size is increased to nearly 12 nm, then $T_{\text{Ntot}} = T = 300$ K, and between $T_{\text{Ntot}}$ and D, there is a linear relationship: $T_{\text{Ntot}} = -440.6 + 83D$. This is a very useful result that can be applied in magnetic devices and in biomedical applications.

Keywords: isothermal entropy; Néel phase transition temperature; synthetic nanomaterials; $\text{Fe}_2\text{O}_3$ thin films; the Monte-Carlo method

1. Introduction

Thin films, two-dimensional physical systems with different structures and materials, have been studied for many decades, especially after the discovery of the so-called giant magnetoresistance effect [1,2] and topological phase transitions [3,4]. Research in this domain not only gives us theoretical results concerning the foundations of modern physics but also leads to the nanotechnology of thin layers, which gives different new functional materials with significant applications in practice. Recently, Prof. Mirosław Dudek et al. [5–9] intensively studied mechanical and magnetic characteristics of different two-dimensional nanocomposite materials. We will discuss their results obtained in detail below. We would like to emphasize that our paper follows this direction of study.

Nowadays, magnetic nanocomposites play an important role in science and technology [10,11]. It is important in practical applications to find materials that can be used in devices such as force sensors and refrigeration equipment. These devices always ensure requirements such as high load capacity, good corrosion resistance, and lightweight. To ensure these requirements, we choose $\text{Fe}_2\text{O}_3$ nano synthetic thin film as the subject of this paper. The reason is that the synthetic nano-$\text{Fe}_2\text{O}_3$ thin film is an antiferromagnetic material [12–17] with many advantages relative to the original material. Moreover, it is widely used in practical applications such as recording equipment [18,19], refrigeration
Coatings 2021, 11, 1209

Wenger et al. [46] have successfully determined that the magnetization (M) of Fe-O bond was approximately 20 kOe. Due to this, Fe₂O₃ is a ferromagnetic material, namely T² approximately 20 kOe. When materials with high or low magnetism are considered, researchers concentrate on the entropy change (ΔS). This quantity is determined through the Maxwell equipment [20,21], printing devices [22], photocatalyst materials [23,24], ion recovery [25], magnetic nanofilm [26], magnetic resonance [27,28], magnetic fluids [29], pigments [30], gas sensors [31,32], the biomedical field [33–37], and spintronics [38,39]. In addition, Fe₂O₃ is also a material with many structures, such as α-Fe₂O₃ (hematite), β-Fe₂O₃ and γ-Fe₂O₃ (magnehite), and ε-Fe₂O₃ [40]. Among them, the α-Fe₂O₃ hematite structure is the most commonly used today, while the ε-Fe₂O₃ structure is the most difficult structure to be manufactured. The ε-Fe₂O₃ structure has not been studied extensively. Applying the experimental method, Zuohui Cheng et al. [41] have successfully determined the influence of enthalpy and entropy on the size of Fe₂O₃ materials. They concluded that when the size (D) increases from D = 19.3 nm to D = 140.5 nm, the total magnetization (Mₜot) increases, and the Neél phase transition temperature (Tₜot) increases from Tₜot = 742 K to Tₜot = 897 K [41]. Liu et al. [42] proposed an approximate expression for the enthalpy and entropy of the nanoparticles without considering the change in temperature during the transition. Cui et al. [43] demonstrated that the crystallinity densities of the two transitions are equal. Zhang et al. [44] successfully determined the Neél phase transition temperature of Fe₂O₃ when considered system transitioned from the maghemite phase to the α hematite phase, namely Tₜot = 623 K at D = 4 nm and Tₜot = 723 K at D = 24 nm. Mendili et al. [45], Wenger et al. [46] have successfully determined that the magnetization (M) of γ-Fe₂O₃ nanoparticles increases when D increases (from D = 2 nm to D = 4 nm). Hou et al. [47] suggested that the coercivity of nano-Fe₂O₃ increases when the D size increases (from D = 12 nm to D = 40 nm). Jun Wang et al. [48] successfully determined the Tₜot of α-Fe₂O₃ as Tₜot = 930 K. In the study of José Luis García-Muñoz [49] for ε-Fe₂O₃, this temperature was Tₜot = 850 K. The obtained results show that, for bulk or nanomaterials, an increase in D leads to an increase in Tₜot. Additionally, it was found that, for Fe₂O₃ synthetic thin film at room temperature T = 300 K, there exists a very large magnetic force of approximately 20 kOe. Due to this, Fe₂O₃ materials are used frequently in high-density magnetic recording devices, high-frequency electromagnetic waves absorbers [50–52]. On the other hand, ε-Fe₂O₃ is a ferromagnetic material [53]. In some kind, it is an intermediate form between γ-Fe₂O₃ and α-Fe₂O₃, so its Neél phase transition temperature ranges from Tₜot = 150 K to Tₜot = 500 K. It has a lowest temperature of Tₜot = 150 K [54,55], and it is characterized by a decrease in coercivity and saturation magnetization [54]. In addition, at the cooling condition of Fe₂O₃, the Tₜot value is in the range of Tₜot = 85–150 K, when there is a change in spins in the rhombohedral structure and a non-monotonic change in the Fe-O bond [49,56], even though the Curie temperature (Tₜot) of ε-Fe₂O₃ has a value of Tₜot ~ 500 K [57–60]. Additionally, magnetic ε-Fe₂O₃ nanoparticles can also be obtained by vibrating magnetometer, which shows Tₜot > 500 K and a maximum value of Tₜot = 850 K. The simulation method has received great attention, because researchers can study materials in extreme conditions, such as at high temperature (T), T = 700 K, pressure (P), P = 360 GPa, and with atomic size below 2 nm, where the experimental methods cannot be applied [61–63]. Meanwhile, the magnetic refrigeration technology industry attracts great attention from researchers. Cooling technology is based on the magnetic effect of materials, which was first discovered by E. Warburg in 1881 [64]. When the material is placed in the magnetic field (B), under the action of the magnetic field, the spins rotate in the direction of the magnetic field, leading to the total magnetic (Mₜot) and total entropy (Sₜot) change (increase or decrease), depending on the nature of magnetic materials. In the field of refrigeration technology at room temperature T = 300 K, researchers often use soft magnetic materials which operate at room temperatures, such as Gadolinium with Tₜot = 296 K. It is the first used material with very high cost, poor oxidation resistance, and low magnetic value. For that, researchers try to find materials with high oxidation resistance and low cost, which exhibit high magnetic properties that are essential for future applications. Presently, materials such as transition metals or rare earth materials are still considered materials with great potential for refrigeration applications at room temperature [65–71]. When materials with high or low magnetism are considered, researchers concentrate on the entropy change (ΔS). This quantity is determined through the Maxwell
relationship and the isothermal magnetization curve. However, up to now, the use of the Maxwell relationship to determine entropy change is still causing much controversy and discussions about it [72–76]. Giguère et al. [72] successfully determined the entropy change based on the Maxwell relationship. Balli et al. [75] and Liu et al. [76] also successfully demonstrated that the Maxwell relationship no longer exists when the material is near the phase transition (paramagnetic phase, ferromagnetic phase, antiferromagnetic). They suggested that the cause of the appearance of the giant magnetic field at room temperature is due to the morphological leading to changes in the magnetic transition temperature at the T~110 K. In addition, there is an arrangement of the magnets. Different cations lead to a structural change from tetrahedral to octahedral, driven by spin-oriented ions at the magnetic particles, and a magnetic transition occurs at T_{Ntot}~150 K. T. Muto et al. [77] studied the entropy, whereas P. Fratzl [78] and Dieter [79] determined the phase transition diagram. In 2010, Jiri Tucek et al. [80] successfully determined the existence of the huge magnetic field of nano ε-Fe$_2$O$_3$ at room temperature. Recently, M.R. Dudek et al. have successfully determined the giant magnetic field of Fe$_3$O$_4$ nanomaterials at room temperature [6], the magnetic domain in auxetic materials [7,8], and successfully constructed a single-body Hamiltonian function [6,12,17]. Combining with the phase space average field in [9] to study the magnetism of Fe$_3$O$_4$ nanoparticles [81], Dung et al. [82] determined the magnetic properties of Fe nanomaterials by the Monte-Carlo simulation method with the classical Heisenberg model. In addition, researchers also successfully studied the influence of factors such as temperature, number atomic, pressure, annealing time on structure, electronic structure, phase transition, and crystallization progress of material metals [83–87], alloys [88–92], oxide [61–63], and polymers [93,94]. Here, a question appears: how to determine the magnetic characteristic quantities of materials such as magnetization in all directions and entropy of the material when the size of the material is less than 10 nm. To answer this question, in this article, we focus on a study of the influence of factors such as material size (mxm), magnetic field (B), and nanoparticle size on magnetization in the z-axis (M$_z$), total composite magnetization (M$_{tot}$), and total entropy of Fe$_2$O$_3$ nano synthetic thin films. For this purpose, we used the Monte-Carlo simulation method. The obtained results will serve as a basis for future experimental studies when we try to apply Fe$_2$O$_3$ nano synthetic thin films to smart devices and refrigeration equipment.

2. Method of Calculation

Initially, the two-dimensional model for Fe$_2$O$_3$ nano synthetic thin film is constructed by creating a 2D matrix square (mxm). These matrices are composed of nonmagnetic squares and linked together by hinges defined as the intersection points between the corners of the squares (red color in the figure), and these 2D square matrices may be deformed [95]. Then, spherical Fe$_2$O$_3$ nanoparticles were put into the 2D matrix square. For simplicity, we treat each nanoparticle as a magnetic spin. When the thin film model is not deformed, the rotation angle of these 2D square matrices has the value $\alpha = 0^\circ$ (Figure 1a). When the 2D square matrices rotate, the corresponding rotation angle varies from $\alpha = 0^\circ$ to $\alpha = 90^\circ$ (Figure 1b).

In this model, the size of each 2D square is the diameter of the inserted Fe$_2$O$_3$ nanoparticle, with D = 2R, where R is the radius of the nanoparticle. We studied the magnetic properties of Fe$_2$O$_3$ nano synthetic thin films by applying a potential force field to the nano synthetic thin film, with the value of the Hamilton function of the form (Equation (1)) [6], and numerical simulation was performed by the Monte-Carlo method.

$$H_{mfa}^{(i)} = -K_a V \cos^2 \alpha_i - BM \cos \alpha_i - \sum_{j} K_{ij} \mathbf{m}_i \cdot \langle \mathbf{m}_j \rangle$$

where $K_a = \frac{1}{k_B T} \frac{\mu_0}{2 \pi M}$, $V = \frac{4}{3} \pi a_0^3$, $a_0 = 8.394$ Å, and $K_{ij} = \frac{\mu_0 M^2}{4 \pi d}$, $d = \sqrt{2} \sin(\alpha + \frac{\pi}{4})$, $a = D + a_0$, $D = 2K_g$. 

$$M_z = \frac{\mu_0 M^2}{4 \pi d} \cdot \frac{V}{\left(\frac{4}{3} \pi a_0^3\right)}$$

and $K_g = \frac{1}{k_B T} \frac{\mu_0}{2 \pi M}$.
The initial shape of materials has a square structure with a rotation angle $\alpha = 0^\circ$ (a), rotation angle $\alpha \neq 0^\circ$ (b).

In it, $K_a$ is the uniaxial magnetic anisotropy energy; the magnetic coefficient is $\mu_0 = 4\pi \times 10^{-7}$ m/A; $V$ is the volume of the nanoparticle; $a_0$ is the lattice constant; $K_{ij}$ is the interaction energy between the $i$th nanoparticle and the nearest $j$th nanoparticle; and Boltzmann’s constant $k_B = 1.38 \times 10^{-23}$ J/K = 8.617 × 10⁻⁵ eV/K, where $B$ is the magnetic field, $M$ is magnetic moment, $\alpha_i$ is the rotation angle of the $i$th 2D square matrix, $m_i$ and $m_j$ are the magnetic moments of the $i$th and $j$th atom, and $d$, $a$, $R_g$, and $D$ are the distance between the centers of the two nearest nanoparticles, the size of the square edge, the displacement radius, and the size of the Fe$_2$O$_3$ nanoparticle, respectively. Conversely, the size of the model is determined by the following formula (Equation (2)):

$$L = (m - 1) \times d$$  \hspace{1cm} (2)

Here, $m$ is the number of rows (columns) of the matrix. The interaction between the Fe$_2$O$_3$ nanoparticles was determined by the magnetic dipole interaction. Then, Fe$_2$O$_3$ nanoparticles are affected by magnetic moments in all directions.

In the spherical coordinate system: $m_x = \sin \alpha \cos \varphi$, $m_y = \sin \alpha \sin \varphi$, $m_z = \cos \alpha$; $0 < \alpha < 180^\circ$, $0 < \varphi < 360^\circ$. The total magnetic moment ($M_{\text{tot}}$) is expressed by the following expression (Equation (3)) [6]:

$$M_{\text{tot}} = \sqrt{M_x^2 + M_y^2 + M_z^2} = \pm 1; \quad M_{\text{tot}} = \frac{1}{N} \sum_{i=1}^{N} S_i, \quad S_i = \pm 1, \hspace{1cm} (3)$$

where $S_i = +1$ with spin up, and $S_i = -1$ with spin down of Fe$_2$O$_3$ nanoparticles.

In the nano synthetic thin film with the size $L$, $V_s = b(Ld)^2$ is the thin film body and $b$, $d$, $\alpha$, and $\varphi$ are, respectively, the nanoparticle thickness, the distance between the nanoparticles, the polarization angle (pointing the direction of the magnetic moment in the $x$-$y$ plane), and the azimuth (pointing the direction of the magnetic moment for the $z$-axis).

To study the magnetic properties of Fe$_2$O$_3$ nano synthesized thin films, various authors have applied the Monte Carlo method in numerical simulations [96–98]. To increase the accuracy of the results, we used periodic boundary conditions to eliminate surface effects. The obtained results are also compared with the results of the density functional theory method to increase the accuracy of the obtained results.

Ising’s 2D model is placed in the magnetic field $B = 0.1$ T while the influencing factors, such as model size (mxm), rotation angle ($\alpha$), and the external magnetic field (B), are changed. To simulate numerically, we used the Metropolis algorithm in the framework of the Monte-Carlo method and surveyed magnetic characteristic quantities in temperatures...
from $T = 0$ to 600 K, with a total number of MC simulation steps $5 \times 10^4$ corresponding to 84 MC steps for each temperature $T = 1$ K. It has been emphasized in [6] that the Monte-Carlo Metropolis code becomes stable very quickly. It follows from Figure 3c of this paper that, from the vicinity of room temperature to the larger temperatures, the results obtained after 20 Monte Carlo steps (MCS) are practically the same as those after 200 MCS. Our calculations show that, after 30 MCS, the change in obtained results is negligible, but for certainty, 84 MCS have been performed.

The simulation method is based on a random generation of energy variation of the system. Next, we rotated their magnetic moment from $\vec{m} = (m_x, m_y, m_z)$ to $\vec{m}' = (m'_x, m'_y, m'_z)$ and calculated the energy values $H_m, H_m'$ with the corresponding probability distribution (Equation (4)) [6]:

$$P(E) = \frac{\exp(-\beta \Delta E)}{Z}, \quad Z = \sum_i^n \exp(-\beta \Delta E)$$

where $P(E)$ is the probability value of finding spin min $(1, \exp(-\beta \Delta E))$ in a state; $\beta = 1/k_B T$; $T$ is the temperature, $Z$ is the partition function, $\Delta E$ is energy variation of the system generated randomly.

To analyze the model, we calculate the magnetic characteristic quantities of the considered system, such as the total entropy (Equation (5)) [6] of the Fe$_2$O$_3$ nano synthetic thin film with the following expression:

$$S_{tot} = \frac{1}{Nk_B} \sum_i^N S_i$$

To determine the Néel phase transition temperature ($T_{Ntot}$) of Fe$_2$O$_3$ nano synthetic thin films, the intersection between the magnetization curve with the entropy curve is fixed. The entire numerical simulation was carried out based on the Python programming code provided by Prof. M.R. Dudek [6]. This code was properly modified for our purpose and was applied on the computational server system of the Institute of Physics, Department of Physics and Astronomy, Zielona Gora University, Poland.

3. Results and Discussion
3.1. Magnetic Characteristic Quantities

We determine the magnetic characteristic quantities of Fe$_2$O$_3$ nano synthesized thin films, such as the preferred magnetization in the z-axis ($M_{zE}$), the magnetization in the z-axis ($M_z$), the total magnetization ($M_{tot}$), total entropy ($S_{tot}$), and Néel phase transition temperature ($T_{Ntot}$).

The Néel phase transition temperature is the phase transition temperature of a material from an antiferromagnetic state to a superparamagnetic state. To determine the characteristic quantities, as has been emphasized above, we treat each spherical nanoparticle as a spin (with $D = 6$ nm and the magnetic moment is determined by Equation (3)). The results are shown in Figure 2.

The results show that when the Fe$_2$O$_3$ nano synthetic thin film is placed in the magnetic field ($B$), $B = 0.1$ Tesla (T), and the spin of the nanoparticles is rotated by an angle $\alpha = 90^\circ$, the shape of the synthesized thin film nano Fe$_2$O$_3$ with $m_xm = 5 \times 5$, nano size ($D$), $D = 6$ nm corresponding to the size $L = 27$ nm is given in Figure 2a. The relationship between the magnetization oriented in the direction of the z-axis ($M_{zE}$) is shown by the black line in Figure 2b; magnetization in the z-axis ($M_z$) is shown by the green line in Figure 2c. Synthetic magnetization of Fe$_2$O$_3$ materials is given by dark blue line in Figure 2d and synthetic entropy ($S_{tot}$) is drawn in red color when the temperature increases.
The Néel phase transition temperature is the phase transition temperature of a material from an antiferromagnetic state to a superparamagnetic state. To determine the characteristic quantities, as has been emphasized above, we treat each spherical nanoparticle as a spin (with \( D = 6 \) nm and the magnetic moment is determined by Equation (3)). The results are shown in Figure 2.

Figure 2. Shape (a), the relationship between magnetic characteristic quantities with different temperatures of Fe\(_2\)O\(_3\) nanocomposite thin film with \( mxm = 5 \times 5 \) (\( L = 27 \) nm), nano size (D), \( D = 6 \) nm, magnetic field B = 0.1 T, and rotation angle \( \alpha = 90^\circ \). The magnetization in the preferred \( z \)-axis (b), the magnetization in the \( z \)-axis (c), and the total magnetization (d) with the total entropy.

Dudek et al. [6] successfully determined the Néel phase transition temperature of the Fe\(_3\)O\(_4\) nano synthetic thin film and showed that the cause of this phenomenon is due to the magnetic effect of the spins. For this reason, we omit the determination of magnetic characteristic quantities such as magnetization (M), specific heat (\( C_v \)), magnetic susceptibility (\( \chi \)), and energy (E) of the thin film synthesized Fe\(_2\)O\(_3\) nano and only focus our attention on studying the relationship between the characteristics of the magnetization M (\( M_{zE} \), \( M_z \), \( M_{\text{tot}} \)) with the total entropy (\( S_{\text{tot}} \)) when the temperature (T) increases (what it has been demonstrated above in Figure 2). The total entropy is determined according to the formula (5). At \( T = 10 \) K, \( M_{zE} = 0.886, M_z = 0.941, M_{\text{tot}} = 0.941, S_{\text{tot}} = -0.069 \), when T increases from \( T = 10 \) K to \( T = 600 \) K with the temperature shift \( dT = 5 \) K, all values of magnetization M (\( M_{zE}, M_z, M_{\text{tot}} \)) decrease as \( M_{zE} \) decreases from \( M_{zE} = 0.886 \) to \( M_{zE} = 0.156 \), \( M_z \) decreases from \( M_z = 0.941 \) to \( M_z = 0.390 \), and \( M_{\text{tot}} \) decreases from \( M_{\text{tot}} = 0.941 \) to \( M_{\text{tot}} = 0.394 \), which leads to an increase in \( S_{\text{tot}} \) from \( S_{\text{tot}} = -0.069 \) to \( S_{\text{tot}} = 2.354 \). The displacement of the spins corresponds to the probability of finding the existence of spins in a given state at a certain temperature. The lines of the magnetization \( M_{zE}, M_z, M_{\text{tot}} \), and the total entropy \( S_{\text{tot}} \) intersect at a point, which is called the magnetic phase transition point or the Néel phase transition temperature (\( T_{N\text{tot}} \)). The intersection between \( M_{zE} \) and \( S_{\text{tot}} \) is \( T_{NzE} = 60 \) K;
between $M_z$ and $S_{tot}$ is $T_{Nz} = 68$ K; and between $M_{tot}$ and $S_{tot}$ is $T_{Ntot} = 68$ K. This is the Néel phase transition temperature from the antiferromagnetic state to the superparamagnetic state. This result is completely consistent with the magnetic effect results previously obtained with Fe$_3$O$_4$ nano synthetic thin films at room temperature [6]. To confirm that, we study the factors affecting the isothermal entropy and Néel temperature of Fe$_2$O$_3$ nano synthesized thin films with $D = 6$ nm.

3.2. The Influence of Different Factors

3.2.1. Effect of the Synthetic Thin-Films

Effect of the Synthetic Thin Film Size

To study the effect (mxm) of Fe$_2$O$_3$ nano synthetic thin films, the size model increases from $mxm = 5 \times 5$ (L = 27 nm) to $mxm = 10 \times 10$ (L = 62 nm), $15 \times 15$ (L = 96 nm), $20 \times 20$ (L = 130 nm), $30 \times 30$ (L = 198 nm), $40 \times 40$ (L = 267 nm) with $D = 6$ nm. The considered system is placed in a magnetic field ($B$), $B = 0.1$ T with a rotation angle of $\alpha = 90^\circ$. The result obtained is shown in Figure 3.

The obtained results show that the Fe$_2$O$_3$ nano synthetic thin film of size $L = 27$ nm has the shape given in Figure 3a. The total magnetization ($M_{tot}$) is shown by the blue line in Figure 3b, and the total entropy ($S_{tot}$) is shown by the red line in Figure 3c. When temperature ($T$) increases from $T = 10$ K to $T = 600$ K, the magnetization ($M_{tot}$) decreases from $M_{tot} = 0.941$ to $M_{tot} = 0.394$, $S_{tot}$ increases from $S_{tot} = -0.069$ to $S_{tot} = 2.354$, and
Neel’s magnetic phase transition temperature ($T_{Ntot}$) increases slightly from $T_{Ntot} = 68$ K to $T_{Ntot} = 68, 68, 71, 73$ K (Figure 3d). The reason for these changes is that increasing temperature $T$ leads to a shift of the domain walls. When the thin film size increases from $L = 27$ nm to $L = 62, 96, 130, 198, 267$ nm, the $M_{tot}$ increases slightly from $M_{tot} = 0.941$ to $M_{tot} = 0.944, 0.947, 0.952, 0.972, 0.983$, respectively, because the increase in the thin film size of $L$ leads to an increase in the density of spins. The obtained results are completely consistent with the simulation results of amorphous Fe nanoparticles [82]. The cause of this phenomenon is due to the size effect (when increasing the lattice size $L$ leads to an increase in $T_{Ntot}$) with a negligible increase in results (about 9%). We chose Fe$_2$O$_3$ nano synthetic thin film with the nano size $D = 6$ nm, mm = 20 × 20 corresponding to the size $L = 130$ nm as standard to study other influencing factors. Further, we investigated the influence of the spin angle of spin on the magnetic characteristic quantities.

Effect of the Spins Rotation Angle

Similarly, as in the case of analyzing the effect of synthetic thin film size, we consider the influence of spin angle using Fe$_2$O$_3$ nano synthetic thin film with $L = 130$ nm ($D = 6$ nm), $B = 0.1$ T, with an angle $\alpha$ that changes from $\alpha = 0^\circ$ to $\alpha = 90^\circ$. The obtained results are shown in Figure 4.

The results show that when the Fe$_2$O$_3$ nano synthetic thin film of size $L = 130$ nm is placed in a magnetic field ($B$) with $B = 0.1$ T and the rotation angle ($\alpha$) increases from $\alpha = 0^\circ$ to $\alpha = 30^\circ, 45^\circ, 60^\circ, 90^\circ$, the shape of the thin film is shown as in Figure 4a. The $M_{tot}$ increases slightly from $M_{tot} = 0.939$ to $M_{tot} = 0.940, 0.941, 0.941$, because an increase in the size $L$ leads to an increase in the density of spins, whereas $S_{tot}$ increases slightly from $S_{tot} = −0.249$ to $S_{tot} = −0.133, −0.107, −0.088, −0.076$. This leads to the decrease in magnetic phase transition temperature ($T_{Ntot}$) from $T_{Ntot} = 93$ K to $T_{Ntot} = 86, 75, 70, 68$ K (Figure 4b). The cause of the change in $T_{Ntot}$ is the fact that an increase in the rotation angle leads to an increase in spin spacing (d) and to a decrease in the magnetization $M_{tot}$. The distance between the nanoparticle centers increases $d > a$, and in consequence, total entropy $S_{tot}$ decreases. It follows from obtained results that when $L = 27$ nm increases to $L = 62, 96, 130, 198, 267$ nm, the $T_{Ntot}$ increases from $68$ K to $75$ K, and when the rotation angle increases from $\alpha = 0^\circ$ to $\alpha = 90^\circ$, $T_{Ntot}$ decreases from $T_{Ntot} = 93$ K to $T_{Ntot} = 68$ K with $L = 130$ nm. The increase in size leads only to an insignificant change of $T_{Ntot}$, and the rotation angle of the matrix will be a very convenient parameter for experimental...
studies with different types of materials used to manufacture thin films. Through the research results on the influence of thin films on the magnetic properties of Fe$_2$O$_3$ nano synthetic thin films, we conclude that the influence factor of the thin film is very small, almost negligible. So, a question arises: what causes the increase or decrease in $T_{N\text{tot}}$? To study the influencing factors of Fe$_2$O$_3$ nanoparticles, we chose a thin film with a size $L = 130$ nm with a rotation angle of $\alpha = 90^\circ$. To answer this question, we continued the study of the influence of nanoparticles and the impact factors of the external magnetic field on experiments.

3.2.2. Effect of Fe$_2$O$_3$ Nanoparticles

To study the influence of Fe$_2$O$_3$ nanoparticles, we used again a matrix of size $L = 130$ nm, $D = 6$ nm with a rotation angle of the matrix $\alpha = 90^\circ$.

Effect of the External Magnetic Field

Let us consider the Fe$_2$O$_3$ nano synthetic thin film with nanoparticle size $D = 6$ nm, $L = 130$ nm into the external magnetic field $B$ with different intensities. The obtained results are shown in Figure 5.

![Figure 5. Shape (a), Néel phase transition temperature of Fe$_2$O$_3$ nano synthetic thin film with L = 130 nm, rotation angle $\alpha = 90^\circ$ for different magnetic fields B = 0.1 T (b), B = 0.3 T (c), B = 0.5 T (d), B = 0.7 T (e), and B = 0.9 T (f).](image)

The results show that when Fe$_2$O$_3$ nano synthetic thin film with matrix size $L = 130$ nm, nanoparticle size $D = 6$ nm is placed in a magnetic field (B) with $B = 0.1$ T, the shape of Fe$_2$O$_3$ thin film is as in Figure 5a. The $M_{\text{tot}}$ composite magnetization decreases from $M_{\text{tot}} = 0.941$ to $M_{\text{tot}} = 0.404$, the $S_{\text{tot}}$ composite entropy increases from $S_{\text{tot}} = -0.076$ to $S_{\text{tot}} = 2.353$, and the magnetic phase transition temperature is $T_{N\text{tot}} = 68$ K (Figure 5b). When the external magnetic field increases from $B = 0.1$ T to $B = 0.3$, $0.5$, $0.7$, $0.9$ T, the point above of total magnetization $M_{\text{tot}}$ increases slightly from $M_{\text{tot}} = 0.941$ to $M_{\text{tot}} = 0.943$, the total entropy always increases from $S_{\text{tot}} = -0.076$ to $S_{\text{tot}} = -0.932$, the lower point of $M_{\text{tot}}$ increases again from $M_{\text{tot}} = 0.404$ to $M_{\text{tot}} = 0.754$, $0.844$, $0.881$, $0.889$, and $S_{\text{tot}}$ again decreases from $S_{\text{tot}} = 2.353$, $1.789$, $1.479$, $1.295$, $1.165$. This leads to a decrease in the magnitude of $M$, while the magnitude of $S_{\text{tot}}$ increases. It implies that $S_{\text{tot}}$ tends to shift towards the negative axis,
which leads to a corresponding increase in $T_{N\text{tot}}$: $T_{N\text{tot}} = 68$ K at $B = 0.1$ T (Figure 5b), $T_{N\text{tot}} = 148$ K at $B = 0.3$ T (Figure 5c), $T_{N\text{tot}} = 228$ K at $B = 0.5$ T (Figure 5d), $T_{N\text{tot}} = 300$ K at $B = 0.7$ T (Figure 5e), $T_{N\text{tot}} = 376$ K at $B = 0.9$ T (Figure 5f). The cause of the change in $T_{N\text{tot}}$ is that an increase in $B$ leads to the stronger orientation of the spins in the preferred direction of the magnetic field and they rotate very strongly with a large magnetic field. So, there is another problem: how to increase $T_{N\text{tot}}$ with a small external magnetic field?

Effect of Nanoparticle Size

When nanoparticle size ($D$) increases, we obtain the results shown in Figure 6.

![Figure 6](image.png)

**Figure 6.** The Néel phase transition temperature ($T_{N\text{tot}}$) of Fe$_2$O$_3$ nano synthetic thin film with $m \times m = 20 \times 20$, $L = 130$ nm, $B = 0.1$ T, rotation angle $\alpha = 90^\circ$ for different nanoparticle sizes: the shape (a), the results for the size $D = 6$ nm (b), $D = 8$ nm (c), $D = 10$ nm (d), $D = 12$ nm (e), $D = 14$ nm (f) and $T_{N\text{tot}}$ depends on $D$ (g).

The results show that when Fe$_2$O$_3$ nano synthetic thin film with nanoparticle size $D = 6$ nm ($L = 130$ nm) is placed in a magnetic field ($B$), with $B = 0.1$ T, rotation angle $\alpha = 90^\circ$, the shape of the film is as in Figure 6a. The $M_{\text{tot}}$ composite magnetization decreases from $M_{\text{tot}} = 0.941$ to $M_{\text{tot}} = 0.404$, $S_{\text{tot}}$ composite entropy increases from $S_{\text{tot}} = -0.076$ to
When the size increases from $D = 6$ nm ($L = 130$ nm) to $D = 8, 10, 12, 14$ nm ($L = 168, 206, 244, 282$ nm), then upper point of total magnetization $M_{\text{tot}}$ increases slightly from $M_{\text{tot}} = 0.941$ to $M_{\text{tot}} = 0.983$, the total entropy always increases from $S_{\text{tot}} = -0.076$ to $S_{\text{tot}} = -0.083$, the lower point of $M_{\text{tot}}$ increases again from $M_{\text{tot}} = 0.404$ to $M_{\text{tot}} = 0.876$, $0.929$, $0.940$, $0.935$, and $S_{\text{tot}}$ again decreases from $S_{\text{tot}} = 2.353, 1.281, 0.705, 0.310, 0.003$, which leads to the decrease in magnitude of $M_{\text{tot}}$, whereas the magnitude of $S_{\text{tot}}$ increases. $S_{\text{tot}}$ tends to shift towards the negative axis, leading to an increase in $T_{\text{Ntot}}$: $T_{\text{Ntot}} = 68$ K at $D = 6$ nm, ($L = 130$ nm) (Figure 6b), $T_{\text{Ntot}} = 164$ K at $D = 8$ nm, ($L = 168$ nm) (Figure 6c), $T_{\text{Ntot}} = 320$ K at $D = 10$ nm, ($L = 206$ nm) (Figure 6d), $T_{\text{Ntot}} = 560$ K at $D = 12$ nm, ($L = 244$ nm) (Figure 6e), $T_{\text{Ntot}} \geq 600$ K at $D = 14$ nm, ($L = 282$ nm) (Figure 6f). The $M_{\text{tot}}$ curve and $S_{\text{tot}}$ curve intersect at a point called the point of magnetic phase transition ($T_{\text{Ntot}}$). Thereby, it shows that there is a relationship between $D$ and $T_{\text{Ntot}}$: for $D = 6$ nm ($L = 130$ nm), $T_{\text{Ntot}} = 68$ K; for $D = 8$ nm ($L = 168$ nm), $T_{\text{Ntot}} = 164$ K; for $D = 10$ nm ($L = 206$ nm), $T_{\text{Ntot}} = 320$ K; when $D = 12$ nm ($L = 244$ nm), $T_{\text{Ntot}} = 560$ K; and for $D = 14$ nm ($L = 282$ nm), $T_{\text{Ntot}} > 600$ K. This result is completely consistent with the results obtained in [99] for the magnetic phase transition temperature ($T_{\text{Ntot}}$) of Fe$_3$O$_4$ nanoparticles (as $D$ increases, the $T_{\text{Ntot}}$ increases and reaches a maximum value $T_{\text{Ntot}} = 860$ K). The cause of this phenomenon is due to the size effect. The results show that, as $D$ increases, $L$ increases. $T_{\text{Ntot}}$ increases nearly in a linear manner with $D$ according to the approximated formula $T_{\text{Ntot}} = -440.6 + 83D$ (Figure 6g). Through this formula, researchers can adjust the nanoparticle size and subtract the $B$ field to be suitable for specific applications. For example, one can fabricate a nano synthetic thin-film operating at $T_{\text{Ntot}} = 300$ K with the Earth’s magnetic field. For this purpose, we study below the influence of magnetic field $B$, nanoparticle size $D$ on the $T_{\text{Ntot}}$ at room temperature $T = 300$ K. The results of the influence of $B$ and $D$ on the $N_{\text{tot}}$ show that, when increasing both $B$ and $D$, we have a decrease in $M_{\text{tot}}$ and an increase in $S_{\text{tot}}$. This leads to the conclusion that the magnetization of the material always decreases, and the entropy of materials increases. This is very interesting for future applications of magnetic nanomaterials.

Relationship between $B$ and $D$ at Room Temperature 300 K

Considering the above research results, we investigate the influence at room temperature. We investigate the influence of nanoparticle size at the values of $B = 0.025, 0.045, and 0.065$ T with dimensions $D = 10, 12, and 14$ nm (corresponding to $L = 114, 174, and 234$ nm). The results are shown in Figure 7.

The results show that, when the Fe$_3$O$_4$ nano synthetic thin film is placed in the external magnetic field $B = 0.025$ T with the rotation angle $\alpha = 90^\circ$ for the increase in the size of the nanoparticle $D$ from $D = 10$ nm ($L = 206$ nm) to $D = 12, 14$ nm ($L = 244, 282$ nm), $M_{\text{tot}}$ decreases from $M_{\text{tot}} = 0.943$ to $M_{\text{tot}} = 0.740$, and $S_{\text{tot}}$ increases from $S_{\text{tot}} = -0.597$ to $S_{\text{tot}} = 1.803$; whereas for $D = 10$ nm ($L = 206$ nm), $M_{\text{tot}}$ decreases from $M_{\text{tot}} = 0.941$ to $M_{\text{tot}} = 0.871$, and $S_{\text{tot}}$ increases from $S_{\text{tot}} = -0.875$ to $S_{\text{tot}} = 1.304; with D = 12$ nm ($L = 244$ nm), $M_{\text{tot}}$ decreases from $M_{\text{tot}} = 0.936$ to $M_{\text{tot}} = 0.909$, and $S_{\text{tot}}$ increases from $S_{\text{tot}} = -0.908$ to $S_{\text{tot}} = 1.002$. For $D = 14$ nm ($L = 282$ nm) we also have a decrease in the magnitude of $M_{\text{tot}}$ and an increase in $S_{\text{tot}}$. Therefore, as $B$ and $D$ increase, the magnetic phase transition temperature increases from $T_{\text{Ntot}} = 185$ K (Figure 7a1) to $T_{\text{Ntot}} = 324$ K (Figure 7a2), 515 K (Figure 7a3). For $B = 0.045$ T, when $D$ increases from $D = 10$ nm ($L = 206$ nm) to $D = 12, 14$ nm ($L = 244, 282$ nm), the magnetic phase transition temperature increases from $T_{\text{Ntot}} = 220$ K (Figure 7b1) to $T_{\text{Ntot}} = 383$ K (Figure 7b2), 596 K (Figure 7b3). With $B = 0.065$ T, when $D$ increases from $D = 10$ nm ($L = 206$ nm) to $D = 12, 14$ nm ($L = 244, 282$ nm), the magnetic phase transition temperature increases from $T_{\text{Ntot}} = 255$ K (Figure 7c1) to $T_{\text{Ntot}} = 445$ K (Figure 7c2), and $T_{\text{Ntot}} > 600$ K (Figure 7c3). The obtained results show that, at room temperature, because the magnetic field of the earth, $B$, is very small, one can increase the nanoparticle size to nearly 12 nm, then $T_{\text{Ntot}} = T = 300$ K. In addition, between $T_{\text{Ntot}}$ and $D$ there is a relationship that satisfies the equation $T_{\text{Ntot}} = -440.6 + 83D$. This is a very useful result. In
practice, researchers can manufacture Fe$_2$O$_3$ thin films right at ambient conditions (at room temperature). Achieving the size $D = 12$ nm ($L = 244$ nm), these thin films can be used not only in magnetic devices.

Figure 7. Néel phase transition temperature ($T_{Ntot}$) of Fe$_2$O$_3$ nano synthetic thin film $m 	imes m = 20 \times 20$, in $B = 0.1$ T with rotation angle $\alpha = 90^\circ$, for $D = 10$ nm and different values of $B$, $B = 0.025$ T ($a_1$), $B = 0.045$ T ($b_1$), $B = 0.065$ T ($c_1$); for $D = 12$ nm and different values of $B$, $B = 0.025$ T ($a_2$), $B = 0.045$ T ($b_2$), $B = 0.065$ T ($c_2$); for $D = 14$ nm and different $B$, $B = 0.025$ T ($a_3$), $B = 0.045$ T ($b_3$), $B = 0.065$ T ($c_3$).

4. Conclusions

In this study the following results were obtained:

- We successfully studied the influence of the matrix structure ($m \times m$) of thin-film, rotation angle ($\alpha$), magnetic field ($B$), and size ($D$) of Fe$_2$O$_3$ nanoparticle on the magnetic characteristic quantities such as the magnetization-oriented $z$-direction ($M_{zE}$), $z$-axis magnetization ($M_z$), total magnetization ($M_{tot}$), and total entropy ($S_{tot}$) of Fe$_2$O$_3$ nanocomposites by Monte-Carlo simulation method.

- We successfully determined the magnetic phase transition temperature Néel ($T_{Ntot}$). The obtained results show that when the $m \times m$ increases from $m \times m = 5 \times 5$ ($L = 27$ nm)
to \( m \times m = 10 \times 10, 15 \times 15, 20 \times 20, 30 \times 30, 40 \times 40 \) (\( L = 62, 96, 130, 198, 267 \) nm), the \( T_{N_{\text{tot}}} \) increases slightly from \( T_{N_{\text{tot}}} = 68 \) K to \( T_{N_{\text{tot}}} = 73 \) K. When the matrix rotation angle \( \alpha \) increases from \( \alpha = 0^\circ \) to \( \alpha = 90^\circ \), the \( T_{N_{\text{tot}}} \) decreases slightly from \( T_{N_{\text{tot}}} = 93 \) K to \( T_{N_{\text{tot}}} = 68 \) K. The increase in \( B \) (from \( B = 0.1 \) T to \( B = 0.9 \) T) determines an increase in \( T_{N_{\text{tot}}} \) (from \( T_{N_{\text{tot}}} = 68 \) K to \( T_{N_{\text{tot}}} = 148, 228, 300, 376 \) K). The increase in \( D \) (from \( D = 6 \) nm (\( L = 130 \) nm) to \( D = 8, 10, 12, 14 \) nm (\( L = 168, 206, 244, 282 \) nm)) determines an increase in \( T_{N_{\text{tot}}} \) (from \( T_{N_{\text{tot}}} = 68 \) K to \( T_{N_{\text{tot}}} = 164, 320, 560, \) and higher \( 600 \) K). The results show that when \( B \) and \( D \) increase, \( T_{N_{\text{tot}}} \) increases also.

- In addition, between \( T_{N_{\text{tot}}} \) and \( D \), there is a linear relationship that satisfies the equation \( T_{N_{\text{tot}}} = -440.6 + 83D \). This is a very interesting result that can be used in practical applications from cooling technology.

**Author Contributions:** D.N.T.: Conceptualization, Methodology, Investigation, Validation, Resources, Supervision, Writing—original draft—review and editing, Formal analysis. V.C.L.: Validation, Writing and editing. Ş.T.: Writing—editing. All authors have read and agreed to the published version of the manuscript.

**Funding:** This research received no external funding.

**Institutional Review Board Statement:** Not applicable.

**Informed Consent Statement:** Not applicable.

**Data Availability Statement:** The data that support the findings of this study are available from the corresponding author upon reasonable request.

**Acknowledgments:** We would like to express our deep gratitude to M.R. Dudek for support of our work, especially for giving the simulation program code.

**Conflicts of Interest:** The funders had no role in the design of the study; in the collection, analyses, or interpretation of data; in the writing of the manuscript, or in the decision to publish the results and the authors declare no conflict of interest.

**References**

1. Baibich, M.N.; Broto, J.M.; Fert, A.; Dau, F.N.V.; Petroff, F. Giant magnetoresistance of (001)Fe/(001)Cr magnetic superlattices. *Phys. Rev. Lett.* **1988**, *61*, 2472–2475. [CrossRef]
2. Binash, G.; Grinberg, P.; Saurenbech, F.; Zinn, W. Enhanced magnetoresistance in layered magnetic structures with antiferromagnetic interlayer exchange. *Phys. Rev. B* **1989**, *39*, 4828–4830. [CrossRef] [PubMed]
3. Kosterlitz, J.M.; Thoules, D.J. Ordering, metastability and phase transitions in two-dimensional systems. *J. Phys. C Solid State Phys.* **1973**, *6*, 1181–1203. [CrossRef]
4. Haldane, F.D.M. Nonlinear field theory of large-spin heisenberg antiferromagnets: Semiclassically quantized solitons of the one-dimensional easy-axis neel state. *Phys. Rev. Lett.* **1983**, *50*, 1153–1156. [CrossRef]
5. Dudek, K.K.; Gatt, R.; Dudek, M.R.; Grima, J.N. Controllable hierarchical mechanical metamaterials guided by the hinge design. *Materials* **2021**, *14*, 758. [CrossRef]
6. Dudek, M.R.; Dudek, K.K.; Wolak, W.; Wojciechowski, K.W.; Grima, J.N. Magnetocaloric materials with ultra-small magnetic nanoparticles working at room temperature. *Sci. Rep.* **2019**, *9*, 17607. [CrossRef] [PubMed]
7. Dudek, M.R.; Wojciechowski, K.W.; Grima, J.N.; Caruana-Gauci, R.; Dudek, K.K. Colossal magnetocaloric effect in magneto auxetic systems. *Smart Mater. Struct.* **2015**, *24*, 085027. [CrossRef]
8. Dudek, K.K.; Wolak, W.; Dudek, M.R.; Caruana-Gauci, R.; Gatt, R.; Wojciechowski, K.W.; Grima, J.N. Programmable magnetic domain evolution in magnetic auxetic systems. *Phys. Status Solidi RRL* **2017**, *11*, 1700122. [CrossRef]
9. Dudek, M.R.; Grima, J.N.; Cauchi, R.; Zerafa, C.; Gatt, R.; Zapotoczny, B. Space dependent mean eld approximation modelling. *J. Stat. Phys.* **2014**, *154*, 1508–1515. [CrossRef]
10. Mahbouti, Z.; Ali, M.B.; Moussaoui, H.E.I.; Hamedoun, M.; Marssi, M.E.I.; Kenz, A.E.I.; Benyoussef, A. Structural and magnetic properties of \( \text{Co}_{0.7}\text{Ni}_{0.3}\text{Fe}_{2}\text{O}_{4} \) nanoparticles synthesized by sol–gel method. *Appl. Phys. A* **2016**, *122*, 651–655. [CrossRef]
11. Middey, S.; Jana, S.; Ray, S. Surface spin-glass and exchange bias in \( \text{Sr}_2\text{FeMoO}_6 \) nanoparticle. *J. Appl. Phys.* **2010**, *108*, 043918–043923. [CrossRef]
12. Petracic, O.; Chen, X.; Bedanta, S.; Kleeman, W.; Sahoo, S.; Cardoso, S.; Freitas, P.P. Collective states of interacting ferromagnetic nanoparticles. *J. Magn. Magn. Mater.* **2006**, *300*, 192–197. [CrossRef]
13. Bedanta, S.; Petracic, O.; Kleeman, W. Supermagnetism. In *Handbook of Magnetic Materials*; North Holland: Amsterdam, The Netherlands, 2015; Volume 23, pp. 1–83.
44. Zhang, W.; Xue, Y.; Cui, Z. Effect of size on the structural transition and magnetic properties of nano-CuFe₂O₄. *Ind. Eng. Chem. Res.* 2017, 56, 13760. [CrossRef]

45. Mendili, Y.E.; Bardeau, J.F.; Randrianantoandro, N.; Grasset, F.; Greneche, J.M. Insights into the mechanism related to the phase transition from γ-Fe₂O₃ to α-Fe₂O₃ nanoparticles induced by thermal treatment and laser irradiation. *J. Phys. Chem. C* 2012, 116, 23785. [CrossRef]

46. Wenger, L.E.; Tsoi, G.M.; Vaishnava, P.P.; Senaratne, U.; Buc, E.C.; Ratna, N.R.; Naik, V.M. Magnetic properties of γ-Fe₂O₃ nanoparticles precipitated in alginate hydrogels. *IEEE Trans. Magn.* 2008, 44, 2760. [CrossRef]

47. Hou, D.L.; Nie, X.F.; Shao, S.X.; Lu, P.; Luo, H. Studies on the magnetic anisotropy and the coercivity of granular γ-Fe₂O₃ powders. *Phys. Status Solidi* 2015, 161, 459. [CrossRef]

48. Wang, J.; Wu, W.; Zhao, F.; Zhao, G.M. Suppression of the Neel temperature in hydro thermally synthesized γ-Fe₂O₃ nanoparticles. *Nanotechnology* 2016, 27, 9705–9713. [CrossRef]

49. Muñoz, J.L.G.; Romaguera, A.; Fauth, F.; Nogués, J.; Gich, M. Unveiling a new high-temperature ordered magnetic phase in Fe₂O₃. *Chem. Mater.* 2017, 29, 9705–9713. [CrossRef]

50. Namai, A.; Sakurai, S.; Nakajima, M.; Suemoto, T.; Goto, M.; Sasaki, S.; Ohkoshi, S. Synthesis of an electromagnetic wave absorber for high-speed wireless communication. *J. Am. Chem. Soc.* 2009, 131, 1170–1173. [CrossRef] [PubMed]

51. Dung, N.T. Factors affecting the earth’s surface on heterogeneous dynamics of CaSiO₃·Oₓ. *Condens. Matter Mater. Phys.* 2016, 18, 3889–3897. [CrossRef]

52. Schrader, R.; Buttner, G. A new phase of iron (III)-oxide γ-Fe₂O₃. *Phys. Rep.* 2000, 328, 193–201. [CrossRef]

53. Mendili, Y.E.; Bardeau, J.F.; Randrianantoandro, N.; Grasset, F.; Greneche, J.M. Insights into the mechanism related to the phase transition from γ-Fe₂O₃ to α-Fe₂O₃ nanoparticles induced by thermal treatment and laser irradiation. *J. Phys. Chem. C* 2012, 116, 23785. [CrossRef]

54. Gich, M.; Roig, A.; Frontera, C.; Molins, E.; Sort, J.; Popovici, M.; Nogues, J. Large coercivity and low-temperature magnetic reorientation in γ-Fe₂O₃ nanoparticles. *J. Phys. Condens. Matter* 2006, 17, 667–691. [CrossRef]

55. Tseng, Y.C.; Souza-Neto, N.M.; Haskel, D.; Gich, M.; Frontera, C.; Roig, A.; van Veenendaal, M.; Nogues, J. Nonzero orbital moment in high coercivity γ-Fe₂O₃ and low-temperature collapse of the magnetocrystalline anisotropy. *Phys. Rev. B Condens. Matter Mater. Phys.* 2009, 79, 094404. [CrossRef]

56. Sanchez, J.L.; Serrano, A.; Campo, A.D.; Abuin, M.; Fuente, O.R.; Carmona, N. Sol–gel synthesis and nano–micron size characterization of α-Fe₂O₃ and γ-Fe₂O₃. *J. Phys. Chem. C* 2009, 113, 11235–11238. [CrossRef]

57. Giguère, A.; Földéaki, M.; Gopali, B.R.; Chahine, R.; Bose, T.K.; Frydman, A.; Barclay, J.A. Direct measurement of the “giant” adiabatic temperature change in Gd₃Si₂Ge₂. *Phys. Rev. Lett.* 1999, 83, 2262–2265. [CrossRef]

58. Sun, J.R.; Hu, F.X.; Shen, B.G. Comment on “direct measurement of the ‘giant’ adiabatic temperature change in Gd₃Si₂Ge₂”. *Phys. Rev. Lett.* 2000, 85, 4191. [CrossRef] [PubMed]

59. Giguère, A.; Földéaki, M.; Gopali, B.R.; Chahine, R.; Bose, T.K.; Frydman, A.; Barclay, J.A. Direct measurement of the “giant” adiabatic temperature change in Gd₃Si₂Ge₂. *Phys. Rev. Lett.* 1999, 83, 2262–2265. [CrossRef]

60. Földéaki, M.; Chahine, R.; Bose, T.K.; Frydman, A.; Barclay, J.A. Reply. *Phys. Rev. Lett.* 2000, 85, 4192. [PubMed]
75. Balli, M.; Fruchart, D.; Gignoux, D.; Zach, R. The “colossal” magnetocaloric effect in Mn$_{1-x}$Fe$_2$AsMn$_{1-x}$Fe$_2$As: What are we really measuring? *Appl. Phys. Lett.* **2009**, *95*, 072509. [CrossRef]

76. Liu, G.J.; Sun, J.R.; Shen, J.; Gao, B.; Zhang, H.W.; Hu, F.X.; Shen, B.G. Determination of the entropy changes in the compounds with a first-order magnetic transition. *Appl. Phys. Lett.* **2007**, *90*, 032507. [CrossRef]

77. Muto, T.; Takagi, Y. The theory of order-disorder transitions in alloys. In *Solid State Physics*; Seitz, F., Turnbull, D., Eds.; Springer: Berlin/Heidelberg, Germany, 1955.

78. Fratzl, P.; Penrose, O.; Weinkamer, R.; Zizak, I. Coarsening in the ising model with vacancy dynamics. *Phys. A Stat. Mech. Appl.* **2000**, *279*, 100–109. [CrossRef]

79. Dieter, G.E. *Mechanical Metallurgy*, 3rd ed.; McGraw-Hill Ser. in Materials Science and Engineering; McGraw-Hill: New York, NY, USA; London, UK, 1986; p. 212.

80. Tucek, J.; Zboril, R.; Namai, A.; Ohkoshi, S. ε-Fe$_2$O$_3$: An advanced nanomaterial exhibiting giant coercive field, millimeter-wave ferromagnetic resonance, and magnetoelectric coupling. *Chem. Mater.* **2010**, *22*, 6483–6505. [CrossRef]

81. Fuentes-García, J.A.; Díaz-Can, A.I.; Guillen-Cervantes, A.; Santoyo-Salazar, J. Magnetic domain interactions of Fe$_3$O$_4$ nanoparticles embedded in a SiO$_2$ matrix. *Sci. Rep.* **2018**, *8*, 5096. [CrossRef]

82. Dung, T.N.; Cuong, N.C.; Toan, N.T.; Hung, P.K. Factors on the magnetic properties of the iron nanoparticles by classical Heisenberg model. *Phys. B* **2018**, *532*, 144–148.

83. Dung, N.T.; Van, C.L. Effects of number of atoms, shell thickness, and temperature on the structure of Fe nanoparticles amorphous by molecular dynamics method. *Adv. Cite. Eng.* **2021**, *9976633*, 1–12.

84. Dung, N.T.; Phuong, N.T. Understanding the heterogeneous kinetics of Al nanoparticles by simulations method. *J. Mol. Struct.* **2020**, *1218*, 128498.

85. Dung, N.T. Z-AXIS deformation method to investigate the influence of system size, structure phase transition on mechanical properties of bulk nickel. *Mater. Chem. Phys.* **2020**, *252*, 123275.

86. Hue, D.T.M.; Coman, G.; Hoc, N.Q.; Dung, N.T. Influence of heating rate, temperature, pressure on the structure, and phase transition of amorphous Ni material: A molecular dynamics study. *Helioyin* **2020**, *6*, e05548.

87. Dung, N.T.; Cuong, N.C.; Van, D.Q.; Tuan, T.Q. Study the effects of factors on the structure and phase transition of bulk Ag by molecular dynamics method. *Int. J. Comput. Mater. Sci. Eng.* **2020**, *09*, 2050016.

88. Dung, N.T.; Van, C.L. Factors affecting the depth of the Earth’s surface on the heterogeneous dynamics of Cu$_{1-x}$Ni$_x$ alloy, $x = 0.1, 0.3, 0.5, 0.7, 0.9$ by Molecular Dynamics simulation method. *Mater. Today Commun.* **2021**, *102812*, 1–8.

89. Van, C.L.; Van, D.Q.; Dung, N.T. Ab initio calculations on the structural and electronic properties of AgAu alloys. *ACS Omega* **2020**, *5*, 31391–31397.

90. Dung, N.T.; Phuong, N.T. Molecular dynamic study on factors influencing the structure, phase transition and crystallization process of NiCu$_{612}$ nanoparticle. *Mater. Chem. Phys.* **2020**, *250*, 123075.

91. Dung, N.T.; Cuong, N.C.; Van, D.Q. Study on the effect of doping on lattice constant and electronic structure of bulk AuCu by density functional theory. *J. Multiscale Model.* **2020**, *11*, 2030001.

92. Hoc, N.Q.; Viet, L.H.; Dung, N.T. On the melting of defective FCC interstitial alloy γ-Fe$_3$C under pressure up to 100 Gpa. *J. Electron. Mater.* **2020**, *49*, 910–916. [CrossRef]

93. Vu Quoc, T.; Do Ba, D.; Tran Thi Thu, D.; Nguyen Ngoc, L.; Nguyen Thuy, C.; Vu Thi, H.; Cao Long, V.; Tâlu, S.; Nguyen Trong, D. DFT study on some polythiophenes containing benzod[d]thiazole and benzod[d]oxazole: Structure and band gap. *Des. Monomers Polym.* **2021**, *24*, 274–284. [CrossRef] [PubMed]

94. Vu, Q.T.; Tran, T.T.D.; Nguyen, T.C.; Nguyen, T.V.; Nguyen, H.; Vinh, P.V.; Nguyen-Tri, P. DFT prediction of factors affecting the structural characteristics, the transition temperature and the electronic density of some new conjugated polymers. *Polymers* **2020**, *12*, 1207. [CrossRef]

95. Evans, K.E.; Nkansah, M.A.; Hutchinson, I.J.; Rogers, S.C. Molecular network design. *Nature* **1991**, *353*, 124. [CrossRef]

96. Landau, D.P.; Binder, K. *Introduction. In A Guide to Monte Carlo Simulations in Statistical Physics*, 4th ed.; Cambridge University Press: Cambridge, UK, 2014; pp. 1–6.

97. Schweika, W. *Disordered Alloys: Diffuse Scattering and Monte Carlo Simulations*; Springer: Berlin/Heidelberg, Germany, 1998; pp. 1–3.

98. Huang, C.; Marian, J. A generalized Ising model for studying alloy evolution under irradiation and its use in kinetic Monte Carlo simulations. *J. Phys. Condens. Matter Inst. Phys.* J. **2016**, *28*, 425201. [CrossRef] [PubMed]

99. Sun, C.Q.; Zhong, W.H.; Li, S.; Tay, B.K. Coordination imperfection suppressed phase stability of ferromagnetic, ferroelectric, and superconductive nanosolids. *J. Phys. Chem. B* **2004**, *108*, 1080–1084. [CrossRef]