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# *On the numerical simulations of magnetic nanostructures using Monte Carlo method.*

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# *Introduction*

The miniaturization of electronic devices led to increasing interest in the study of magnetic properties of nanostructures.
For a long time micro-miniaturization of magnetic elements and devices were significantly lagged behind the miniaturization of semiconductor devices.
However, in recent years there have been relevant success in this area. The features of the structure of magnetic materials can affect the magnetic properties of these materials and lead to technological progress.
The research in this field is aimed to predict the magnetic properties of possible new compounds. This is very important for the needs of electronics, requesting magnetic materials with a wide range of magnetic properties:
e.g., materials with the maximum permeability (rectangular hysteresis loop), and materials with low losses in the reversal, materials with high initial permeability, materials with constant permeability over a wide range of magnetic fields (slope of the hysteresis loop).

*D. Alloyeau, C. Ricolleau, C. Mottet, T. Oikawa, C. Langlois, Y. Le Bouar, N. Braidy and A. Loiseau, NATURE MATERIALS , 8 (2009) 940.*
*T. Kaneyoshi/Journal of Magnetism and Magnetic Materials 323 (2011) 1145.*
*Y. Li, T.X. Wang / Journal of Magnetism and Magnetic Materials 322 (2010) 2773.*
*M. Rennhofer et al. / Intermetallics 18 (2010) 2069.*

# *Introduction*

* Permanent nanomagnets with a size below 10nm are at the center of intensive research because of their possible applications as ultrahigh-density magnetic-storage devices. The main physical limitation of such a technology originates from the superparamagnetic behavior of very small nanoparticles, as the magnetization direction reversal induced by thermal fluctuations is incompatible with long-time recording.

* Some recent works in this area are already dealing with new practical designs for hard discs based on a new technologies. One can selectively design alloys that guarantee an acceptable thermal stability of the permanent magnetization in very small volumes, enabling scale reduction of the memory bits in the recording media.

# *Introduction*

- A great number of studies have been conducted on the ferromagnetic nanorings driven by its potential application for magnetic random access memory. In contrast to the circular magnetic disk where the transition from the vortex state to the single-domain state can occur below a threshold value of its radius owing to the high exchange energy of the vortex core, in the magnetic nanoring the vortex state can be stably retained on a similar size scale because of the absence of vortex core.
- Based on the stable vortex state, the magnetic nanoring is expected to be a suitable data storage element or logic element for applications of magnetic electronic devices. The previous results have shown that the magnetic transition of nanorings occurs via the onion state and the vortex state, depending on their sizes (thickness, diameter and width), geometric shapes, and magnetic parameters.

# *Introduction*

## *Applications to Nanomedicine*

Preliminary tests with magnetic nanoparticles can identify and bind to certain cells, which were functionalized, seem to confirm the possibility of using magnetic nanoparticles, carriers of chemotherapeutic agents in the fight against cancer.

The presence of magnetic nanoparticles within the tumor cells allows a significant increase in sensitivity of diagnostic tests such as magnetic resonance tomography, which enable the identification of tumors or tumor metastases with much smaller than the millimeter.

Another possibility is to use these particles to destroy the cells. Using magnetic fields to promote agitation of the magnetic particles contained within the cells, induce an increase in temperature of the medium leading to cell death thermal

*http://pt.wikipedia.org/wiki/Wiki*

# *Introduction*

- From an industrial and fundamental point of view, it is then essential to qualitatively and quantitatively study the relationship between the ordered state and the size of the nanostructures. In the past ten years, the structure and chemical ordering of nanostructures have motivated numerous experimental and theoretical studies.

- The growing interest is directed towards the magnetic properties of materials with a nanostructure, such as nanoscaled thin films, nanoparticles, nanorods, nanotube and nanowires, due to the fact that much attraction is directed to their applications in nanotechnology.

- The main physically interesting phenomena is the qualitative change of properties of  magnetic materials, when their size decreases to a nanometer scale.

# *Theoretical simulational approach*

- We present the results of the Monte Carlo simulations of magnetic nanostructures, which are based on the plane structures with the square unit cell at low temperatures.

- The spin configurations, thermal equilibrium magnetization, magnetic susceptibility and the specific heat are investigated for the nanotubes of different diameters, using armchair or zigzag edges.

- The dipolar interaction, Heisenberg model interaction and also their combination are considered for both ferromagnetic and antiferromagnetic cases.

- It turns out that the magnetic properties of the nanotubes strongly depend on the form of the rolling up (armchair or zigzag).

# *Theoretical simulational approach*

- The long-range dipolar interaction is often ignored in theoretical studies of magnetic properties in bulk materials in view of its very small magnitude compared to the exchange interaction. For nanoscale materials the dipolar interaction should be carefully treated as it may compete with short-range exchange energy due to its long-range character and position dependence.

- The effect of dipolar interaction component strongly manifests itself for the small radius nanotubes, while for the larger radius nanotubes the Heisenberg interaction is always dominating. In the thermodynamic part, we have found that the specific heat is always smaller for the nanotubes with smaller radii.

# *Method of calculations*

* The Metropolis Monte Carlo algorithm enables one to obtain the macro-state equilibrium for a physical system at the given temperature T, starting from some randomly chosen initial micro-state and then proceed by performing a very large number of random transformations of the micro-states, until we arrive at the equilibrium macro-state.

* Monte Carlo (MC) methods refer, in a very general sense, to any simulation of an arbitrary system which uses a computer algorithm explicitly dependent on a series of random numbers.

* MC is particularly important in statistical physics, where systems have a large number of degrees of freedom and quantities of interest, such as thermal averages, cannot be computed exactly. In a system with *many* degrees of freedom, for example, the thermal average of some quantity associated with each microstate of the system in equilibrium at temperature *T*.

*K. Binder (Ed.), Monte Carlo Methods in Statistical Physics, Spriger, New York,1979*

# *Method of calculations*

* In the Metropolis algorithm the first conformation is randomly generated.

* At each point in the construction of the chain of conformations a move is attempted to the current conformation.

* If the difference between the energy of the resulting conformation and the energy of the current conformation, $\Delta E$, is negative (*i.e.* the energy of the resulting conformation is smaller than the energy of the current conformation), then the resulting conformation is accepted and it becomes the new conformation in the chain.

* If $\Delta E$ is positive, however, a (pseudo)random number between 0 and 1, $0<R<1$, is generated and the resulting conformation is only accepted if $e^{-\Delta E/T} > R$ . If $e^{-\Delta E/T} < R$ then the resulting conformation is refused.

*N.A. Metropolis, A.W. Rosenbluth, M.N. Rosenbluth, A.H. Teller, E. Teller, J. Chem. Phys. 21 (1953) 1087*

# *Method of calculations*

- In the present work we investigate the possible influence of the nanotube's diameter and the type of the edge on the magnetic properties of nanotubes, based on the plane structure with the square unit cell. In Fig. 1
- The rolling up corresponding to the armchair nanotubes is indicated by the vector (m,0) while the rolling up corresponding to the zigzag nanotubes is indicated by the vector (m,m).
- After rolling, any nanotube is defined by the pair of integer parameters $(m_1,m_2)$, which describe its circumference vector on the initial plane, that is $\vec{L} = m_1\vec{a}_1 + m_2\vec{a}_2$ where are unit cell's vectors (later we used $a$ as a lattice constant, which we set to unity, $a=1$).

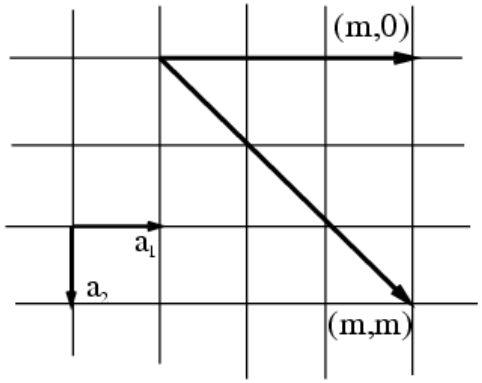



**Figure 1.** Two-dimensional square unit cell lattice. The indicated vectors are used for rolling up the armchair and zigzag nanotubes.

*E. Konstantinova / Journal of Magnetism and Magnetic Materials 320 (2008) 2721*

# *Method of calculations*

We have investigated the spin configurations, the thermal equilibrium magnetization, the susceptibility and the specific heat for these structures.

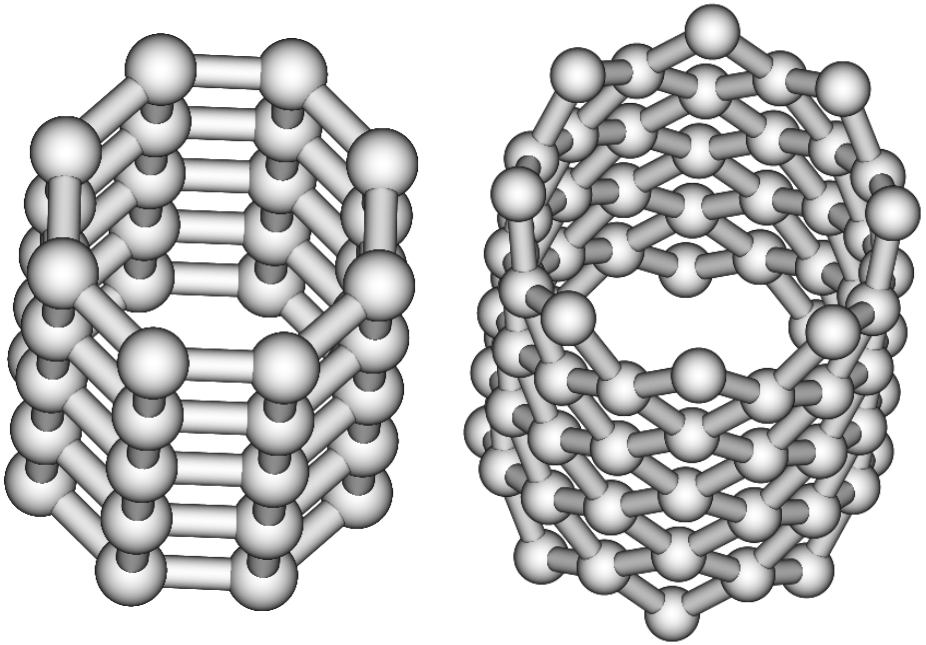


**Figure 2.** Some typical examples of the structures with different edges. The geometries correspond to the armchair (8,0) and zigzag (8,8) nanotubes.

| **Structure** | **Radius** (in units of **a**) |
|---|---|
| (4,0) | 0.6362 |
| (5,0) | 0.7958 |
| (6,0) | 0.9549 |
| (7,0) | 1.1141 |
| (8,0) | 1.2732 |
| (12,0) | 1.9099 |
| (4,4) | 0.9003 |
| (5,5) | 1.1254 |
| (6,6) | 1.3505 |
| (7,7) | 1.5756 |
| (8,8) | 1.8006 |
| (12,12) | 2.7010 |

**Table 1.** The radii of the nanotubes expressed in the units of **a** (unit cell's size).

# *Method of calculations*

In our simulations we used a Hamiltonian model given by

$$H = -J\sum_{\langle i,j\rangle}\vec{S}_i\vec{S}_j - \vec{B}\cdot\sum_i\vec{S}_i - \omega\sum_{i<j}\frac{3\left(\vec{S}_i\cdot\vec{e}_{ij}\right)\left(\vec{e}_{ij}\cdot\vec{S}_j\right)-\vec{S}_i\vec{S}_j}{r_{ij}^3}\cdot$$

The first sum represents the ferromagnetic (or antiferromagnetic) exchange with a coupling constant $J$,

the second sum stands for the coupling of the spins to an external magnetic field $B$,

the last sum is the dipolar interaction term,

where the coupling $\omega$ describes the strength of the dipole-dipole interaction.

The $\vec{S}_i$ are three-dimensional magnetic moments of unit length,

$\vec{e}_{ij}$ are unit vectors pointed from lattice site $i$ to the lattice site $j$,

$r_{ij}$ are the distances between these lattice sites.

The correlation between constant values was chosen as $\omega/J$=0.001, in according to

R. Wieser, U. Nowak, K.D. Usadel, Phys. Rev. B**69** (2004) 064401.

# *Method of calculations*

* For the numerical analysis of the magnetic nanotubes we have used the Monte Carlo simulations with the Metropolis algorithm.

* We start from some randomly chosen initial micro-state and then proceed by performing a very large number of random transformations of the micro-states, until we arrive at the equilibrium macro-state.

* In our case we start simulations with an initial configuration in which all spins have parallel directions. Then the direction of one (randomly chosen) of these spins is randomly changed. In this way we arrive at the new micro and macro-states and evaluate the change of the overall energy $\Delta E$ compared to the previous configuration. If $\Delta E < 0$, the temporary direction of the spin becomes permanent. If $\Delta E > 0$, the temporary direction becomes permanent with the probability $\exp(-\Delta E / k_b T)$.

# *Method of calculations*

* We repeat this procedure $n=10000$ multiplied by the factor equal to the number of sites (spins), because the preliminary calculations show that the equilibrium state configuration is really achieved for $10^4$ Monte Carlo steps per spin and therefore this number of steps is adequate for our calculations.

* In the case of dipolar interaction the spin at the site $i$ was allowed to interact with all others spins of the nanotube.

* The external magnetic field was directed along the axis $z$.

* The simulations for magnetization and magnetic susceptibility were performed $B=-20, -18, \ldots, 0, \ldots, 18, 20$ *(the energy and the applied magnetic field were expressed in units of J, the temperature is expressed in units of* $J/k_b$, *where J is the magnitude of the coupling constant and* $k_b$ *is Boltzmann's constant*).

* In all these cases the value for the temperature was chosen to be $T=0.2$ (*in order to study the low-temperature thermodynamics all simulations have been performed for temperatures essentially smaller than the critical temperature*).

# *Method of calculations*

* We obtain the susceptibility $\chi$ according to the formula

$$\chi = N^{-1}\frac{1}{k_b T}\left(\langle m_z{}^2\rangle - \langle m_z\rangle^2\right),$$

where $N$ is the number of spins in the system

and $\langle m_z\rangle$ is the mean magnetization in the $z$ direction per spin.

* The specific heat $C$ is obtained from the energy fluctuations relation

$$C = N^{-1}\frac{1}{k_b T^2}\left(\langle E^2\rangle - \langle E\rangle^2\right),$$

where $\langle E\rangle$ is energy per spin. For calculating the specific heat

we used $B=0$ and $T=2.0, 1.9, 1.8, 1.7, \ldots, 0.5$.

# *Results*

- Let us start the description of the results from spin orientations for different types of interactions and different values of external magnetic field.
- The simulations have been performed for the longitudinal external magnetic field with the twenty different values of *B* between *-20* and *+20*.
- We start from the B=0 case. In the absence of the external field, considering only the dipolar interaction, we find that the directions of the spins depend on the form of the edge (Fig.3,4).

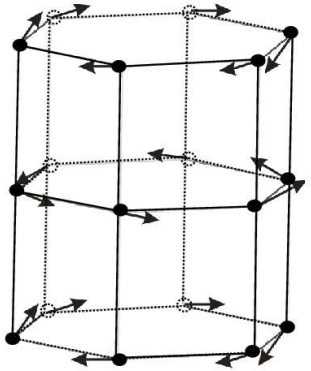


**Figure 3.** The spin configurations for the dipolar interaction without external magnetic field. The illustrative diagram shows the spins in the three layers for the (6,0) nanotube.

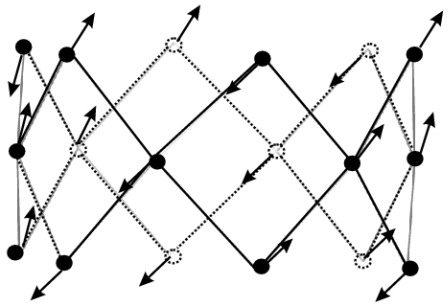


**Figure 4.** The spins distribution for the dipolar interaction without external magnetic field. The illustrative diagram shows the spins in the three layers for the (6,6) nanotube.

# *Results*

- For Heisenberg interaction, the geometric distribution of the spin's directions for the ferromagnetic case is qualitatively the same for all magnetic nanotubes (Fig.5).
- For the Heisenberg-type interaction in the antiferromagnetic cases without the external field the neighbor spins have the opposite directions. For armchair and zigzag type we meet different spin distribution (Fig.6).

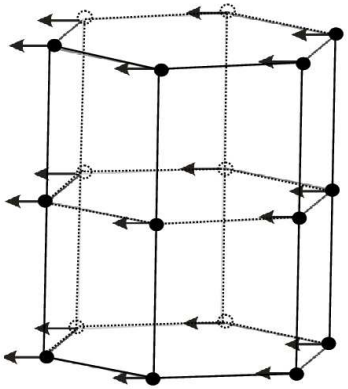


**Figure 5.** These diagrams represent the result for the spin distributions for the Heisenberg interaction in the ferromagnetic case. The example given here corresponds to the three layers for the (6,0) structure.

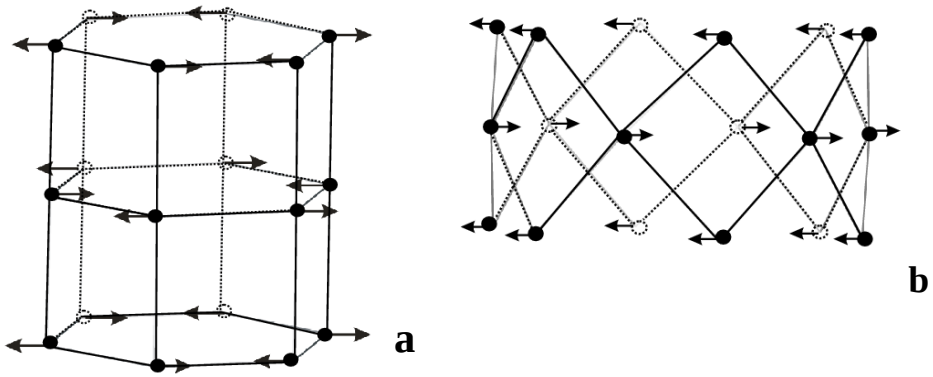



**Figure 6.** These illustrative diagrams represent the result of numerical simulation for the spins distribution for the Heisenberg interaction in the antiferromagnetic case.

* When the dipolar and the Heisenberg interactions are introduced simultaneously, the spin configurations are very similar to the ones for the case of nearest neighboring interactions, as we can see in Figures 5 and 6.

# *Results*

* Now we are in a position to discuss the effect of external magnetic field. In general, the effect of external field is to orient the spins along this field. Of course, this tendency is getting stronger when the external field becomes more intensive, independent on the geometric type of the nanotube.

* For the pure dipolar interaction, the dependences of magnetization on the applied magnetic field have very similar characters for all types of geometric structures under consideration, as it is shown in Figure 7. One can observe certain differences between different nanotubes only for the large values of the applied fields (close to the *B=10).*

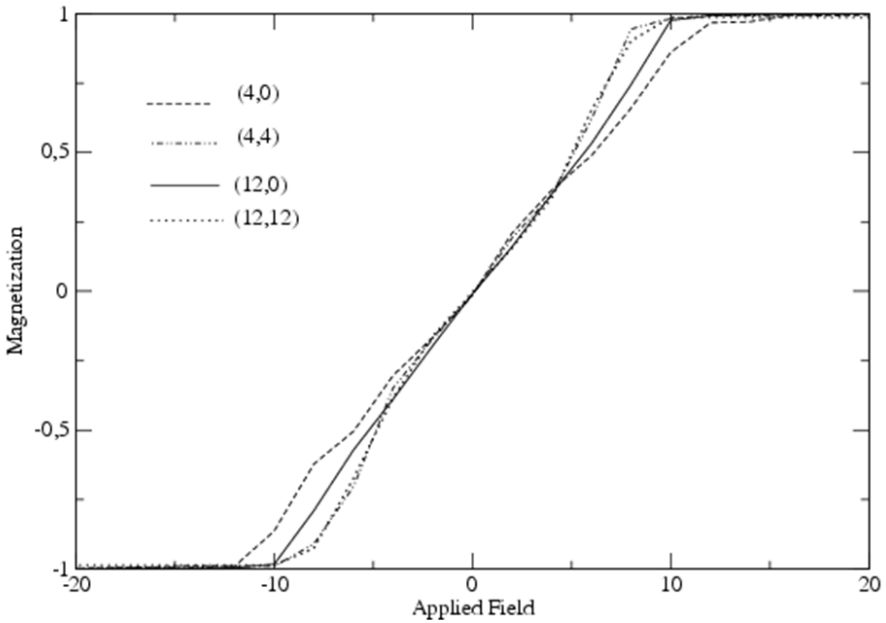



**Figure 7.** The plots of magnetization versus applied field for the dipolar interaction. (4.0) and (12.0) cases have armchair edge while (4.4) and (12.12) represent zigzag edge. In this and next figures the value of the applied magnetic field is in the units of *J*.

# *Results*

* The effect of applied external magnetic field was different for the Heisenberg interaction case between ferromagnetic and antiferromagnetic cases.

* In the antiferromagnetic version for the armchair edge we meet an essentially stronger dependence on the radii of the tubes (Fig.8), in order to achieve the same level of spin orientation, one has to use stronger magnetic field for the thinner tubes. In zigzag case practically does not take place dependence between magnetization and the radius of the nanotube (Fig. 9).

* In the ferromagnetic case, with the Heisenberg interaction, the plots of magnetization versus applied magnetic field are very similar for all radii of the nanotubes and they do not depend of the edge types (Fig.10**)**.

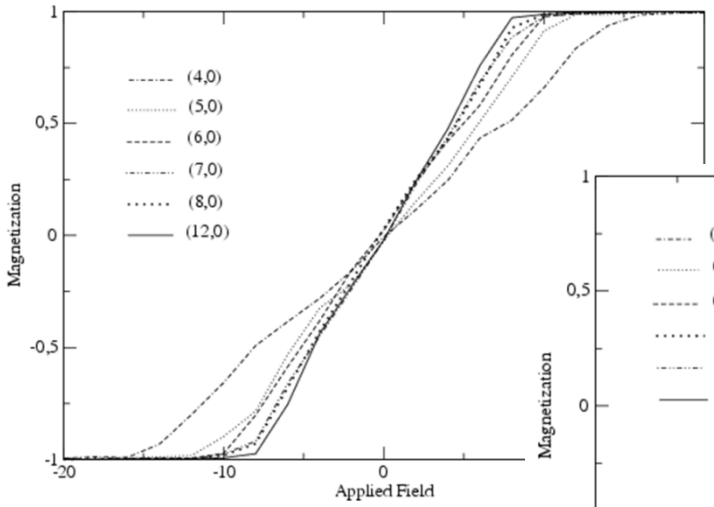



**Figure 8.**

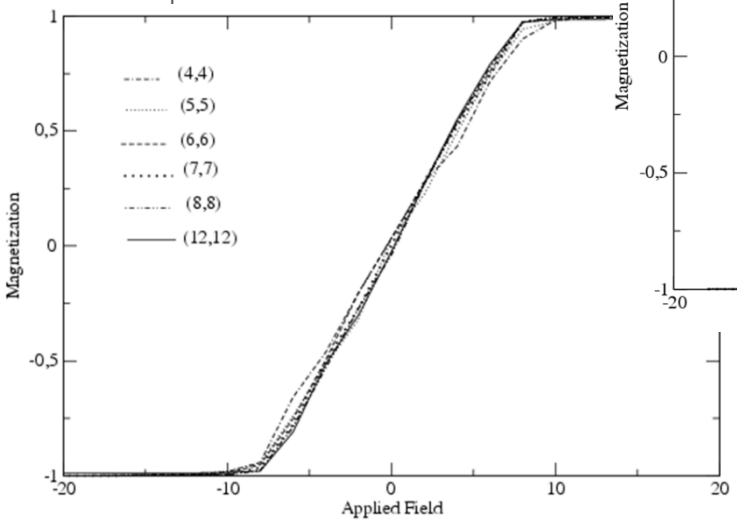



**Figure 9.**

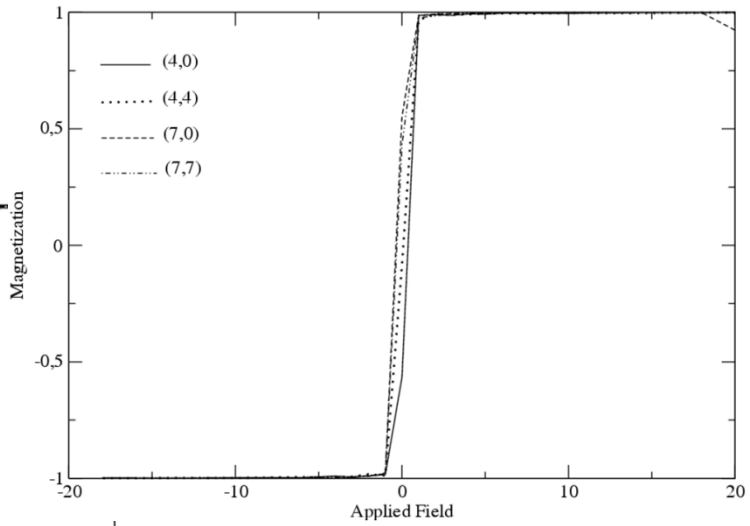



**Figure 10.**

# *Results*

* Finally, let us consider the nanotubes where the dipolar and the Heisenberg-type interactions are introduced simultaneously.

* The plots for the antiferromagnetic case and for the armchair nanotube are in Fig.11. This combination of the two interactions shows stronger dependence on the radius of the tube. For the nanotubes with small diameters we notice that spins orient along of the field very weakly even for the large values of the external field, such that the magnetization remains small even for the maximal field intensity.

* For the zigzag-type nanotubes the plots of magnetizations versus applied magnetic fields are presented in Fig.12 and look rather similar to the pure Heisenberg case. However, for the smallest radius (4,4) tube is an increased influence of the dipolar interaction.

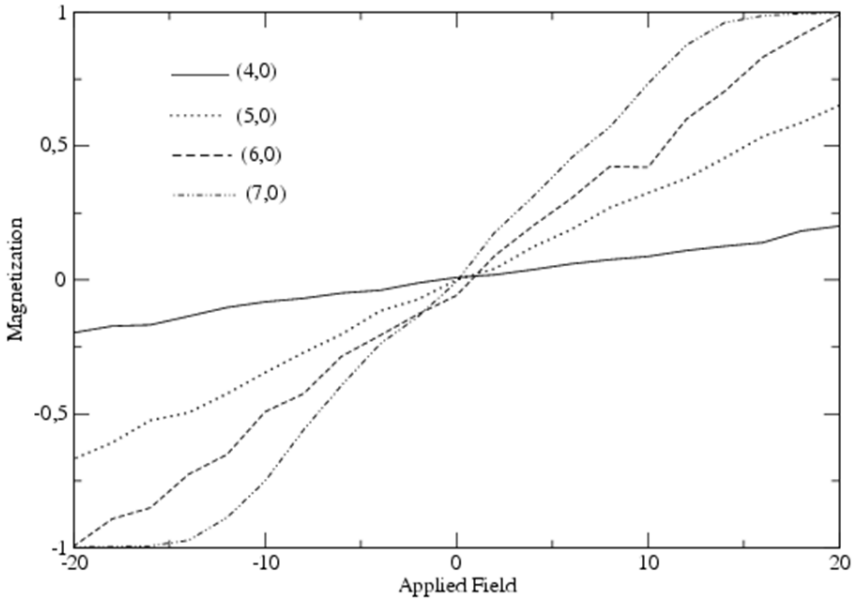



**Figure 11.** Magnetization / applied field for the dipolar and Heisenberg interactions (antiferromagnetic, armchair).

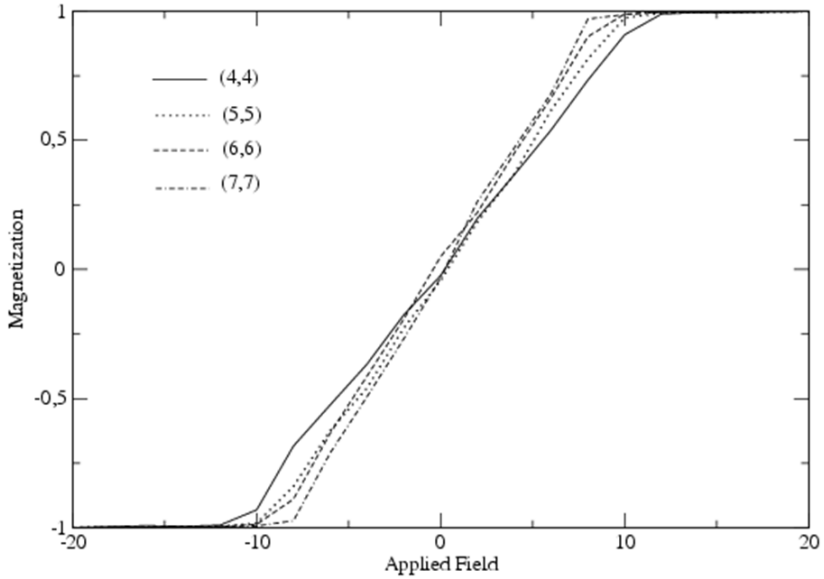



**Figure 12.** Magnetization / applied field for the dipolar and Heisenberg interactions (antiferromagnetic , zigzag).

# *Results*

In Fig.13 we show the similar plots for the structures where the dipolar and the Heisenberg interactions coexist. The dependence of magnetization versus applied field in the ferromagnetic case has the same character as in the Heisenberg interaction. Some influence of the dipolar interactions one can see only for nanotube (4,0).

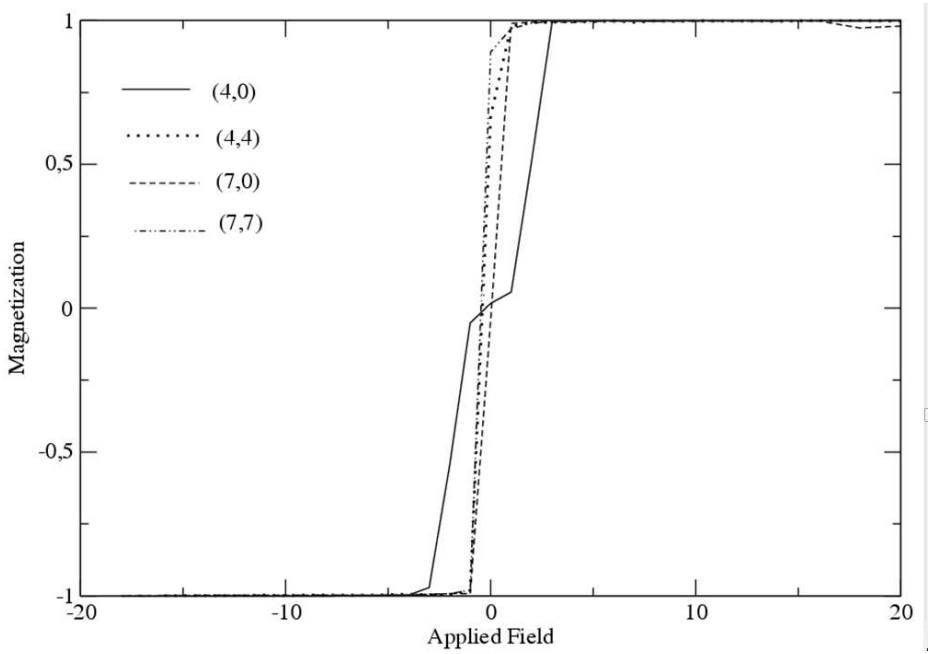



**Figure 13.** The plots of magnetization versus applied field for the sum of the dipolar and the Heisenberg interactions in the ferromagnetic case for armchair and zigzag nanotubes.

# *Results*

- The plots of magnetic susceptibility versus applied field are presented in Figure 14 and 15 for the dipolar interaction cases. The nanotubes for armchair or zigzag edges produce different forms of these plots. One can observe definite similarity between the plots produced for the zigzag edge and different diameters of the tube.

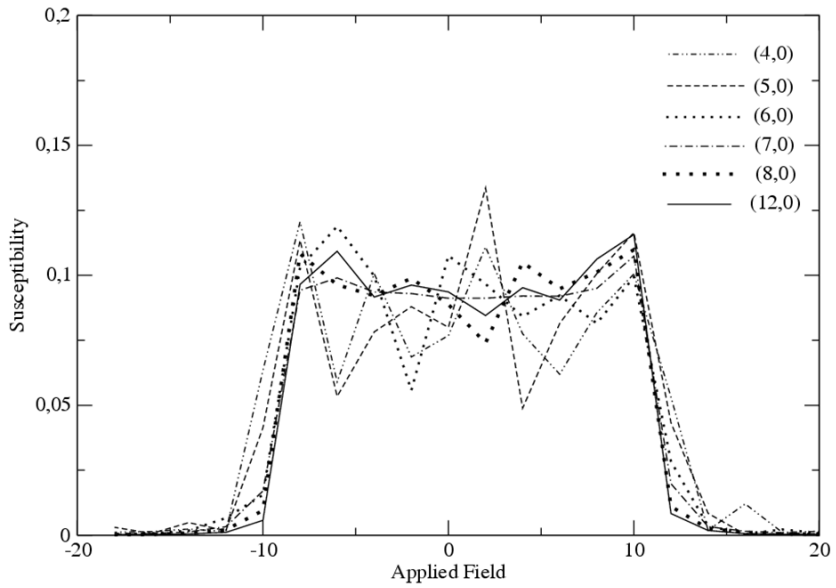



**Figure 14.** Magnetic susceptibility versus applied field for the dipolar interaction for armchair type

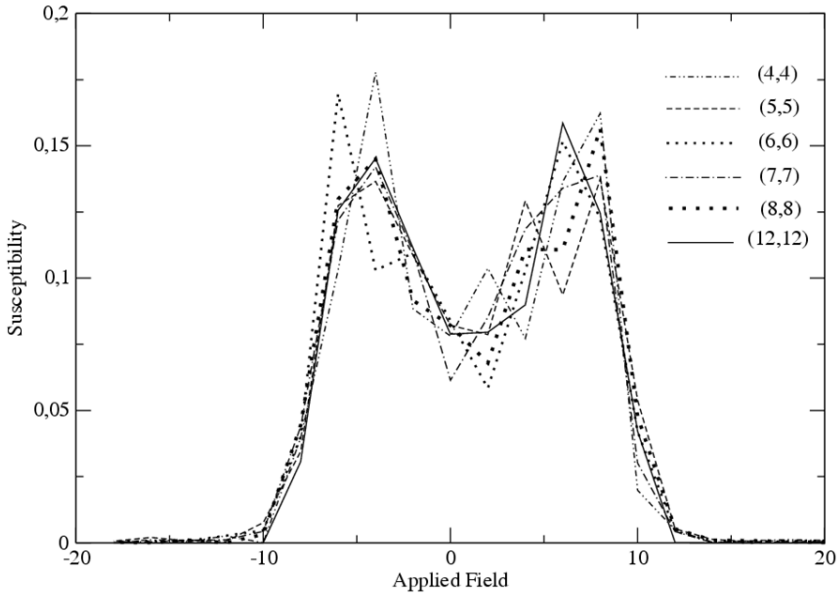



**Figure 15.** Magnetic susceptibility versus applied field for the dipolar interaction in the zigzag type structures.

# *Results*

- For the Heisenberg interaction (antiferromagnetic case) the plots of magnetic susceptibility versus applied field are presented in Figs.16-17. These plots demonstrate stronger dependence on the radius for the armchair edge (Fig.16). The plot looks wider for the smaller radius tubes.
- At the same time, for the zigzag edge (Fig.17) it is difficult to establish such dependence.
- In the ferromagnetic case, with the Heisenberg interaction we meet the same form of the plots for all the structures (Fig.18).

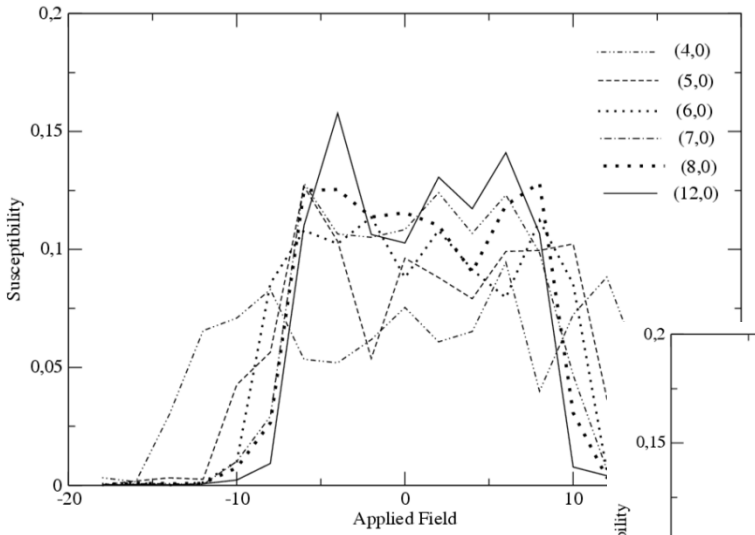



**Figure 16.**

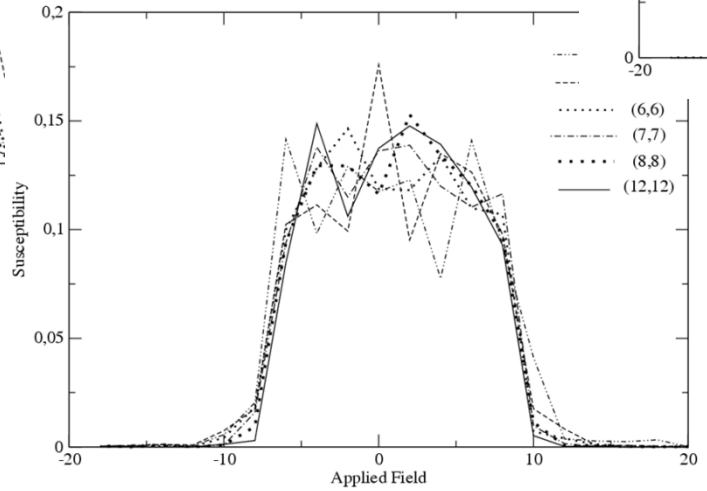



**Figure 17.**

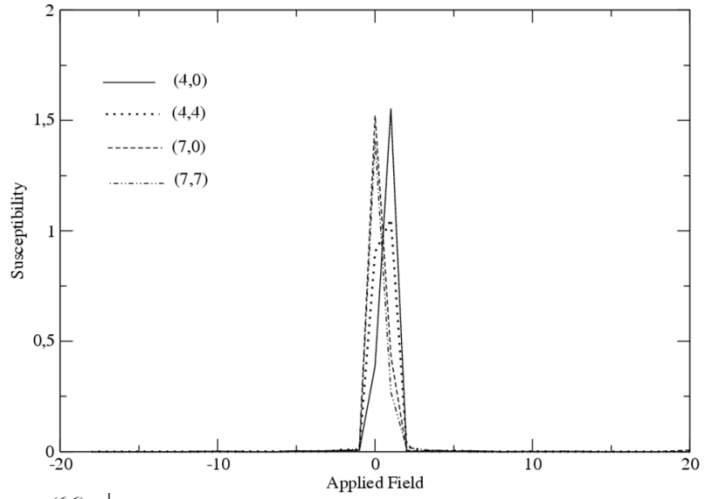



**Figure 18.**

# *Results*

- For the coexisting dipolar and Heisenberg-type interactions, in the antiferromagnetic case (Fig.19) the dependence from the radii of the nanotubes is similar to the one in the Heisenberg-type interaction case. The plot is wider for smaller radius of the tube.
- In the ferromagnetic case for both armchair and zigzag edges we meet wider plots compared to the pure Heisenberg-type interaction (Fig.20), only the (4.0) nanotube gives the plot which is different from the other ones.

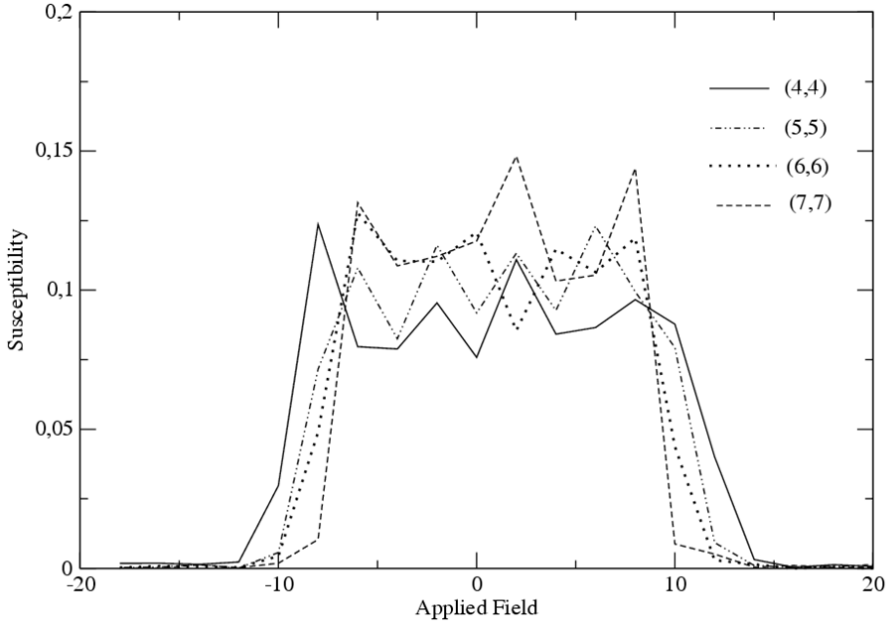



**Figure 19.** Magnetic susceptibility versus applied field for the combination of the dipolar and Heisenberg interactions in the antiferromagnetic case for zigzag nanotubes.

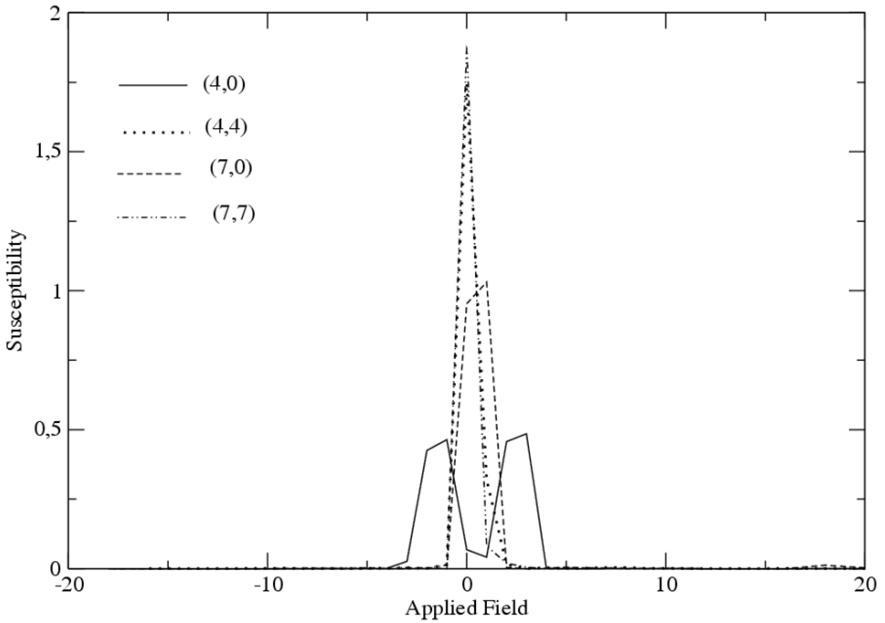



**Figure 20.** Magnetic susceptibility versus applied field for the sum of the dipolar and Heisenberg interactions in the ferromagnetic case for armchair and zigzag nanotubes.

# *Results*

Thermal equilibrium results obtained by Monte Carlo simulations permit us to obtain the specific heat versus temperature.

- For all types of interactions the nanotubes with the smaller radius have smaller value of specific heat.
- In Fig.21 one can notice that the temperature corresponding to the maximal specific heat depends on whether the number of spins in the layer is even or odd.
- In the Heisenberg interactions (Fig.22) the specific heat is smaller for the tubes with smaller radii (the ferromagnetic plots are marked by *).
- For the nanotubes of the same radius, the maximal values of the specific heat correspond to slightly smaller temperatures in the antiferromagnetic case compared to the ferromagnetic case

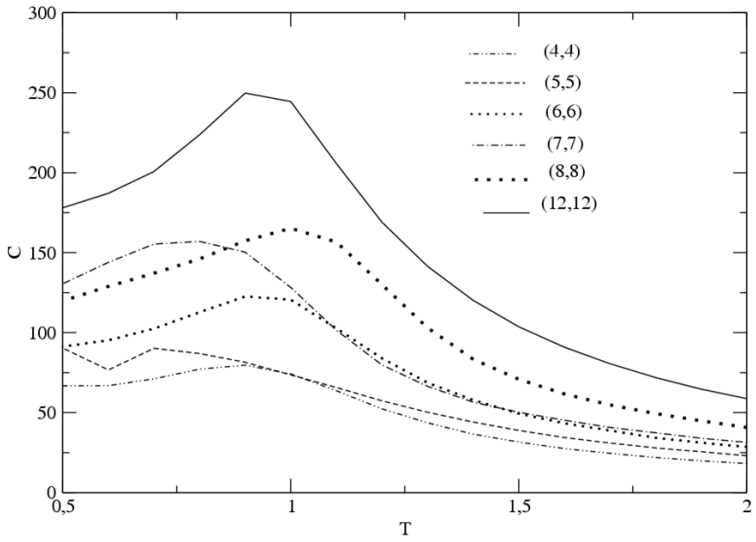
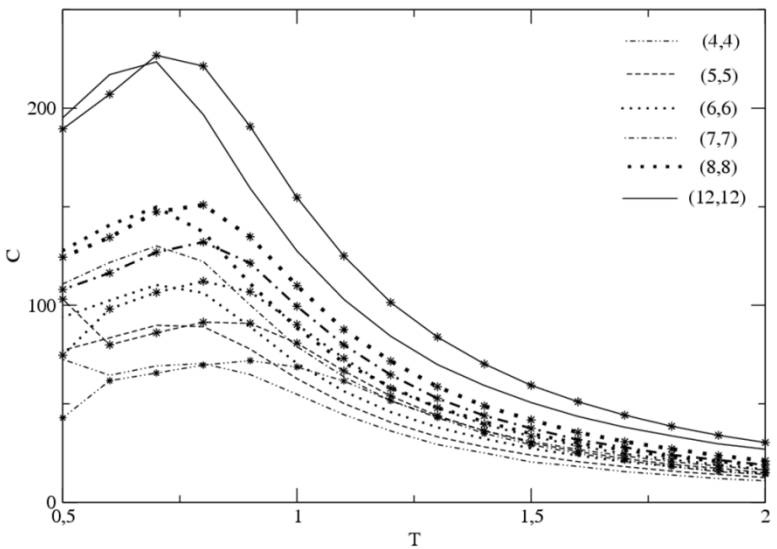



**Figure 21**




In this and next figures the temperature is in units of *J/kb* and the specific heat is in the units of *kb*.

**Figure 22.**

# *Results*

The plots of specific heat versus temperature for the coexisting dipolar and Heisenberg interactions are presented in Figure 23. There is no visible difference between antiferromagnetic and ferromagnetic tubes with the same radii.

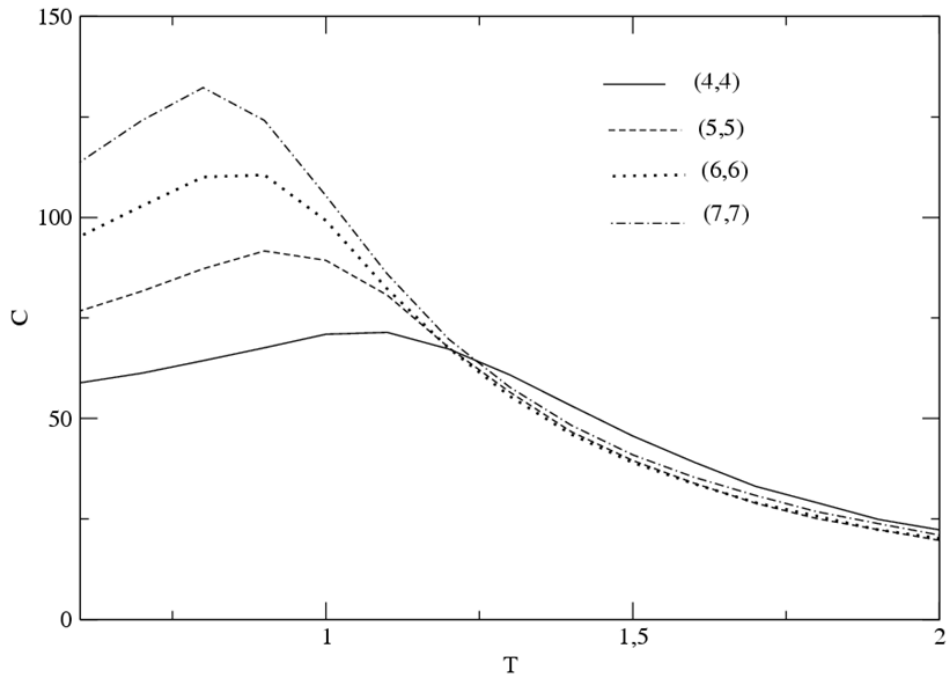



**Figure 23.** The plots of specific heat versus temperature obtained for the combination of the dipolar and Heisenberg interactions in the antiferromagnetic (and ferromagnetic) case for zigzag nanotubes.

# *Results*

•Errors of the calculations strongly depend on the number n. In our case, for the value $n = 10^4$, the errors are very small and they cannot influence the qualitative conclusions concerning the dependence on the geometry, in particular on the diameter of the tube and on the rolling-up rule, which we obtain through the MC simulations.

•In fact, the analysis performed for the selected nanotubes has shown that the degree of the error compared to the value obtained in the calculations does not exceed the value of 0.01.

•This aspect of the theory is well known and has been discussed, for instance, in

*J.F. Fernández, J.J. Alonso, Phys. Rev. B.**76** (2007) 014403.*
*R. Wieser, U. Nowak, K.D. Usadel, Phys. Rev. B**69** (2004) 064401.*
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# *Conclusions*

The magnetic and thermodynamic properties of the nanotubes based on the square form of the unit cell manifest strong dependence on the on their geometry on the form the rolling rule and on the diameter of a nanotube.

This dependence concerns the spin distribution, magnetic susceptibility, magnetization and specific heat. In particular, the specific heat is always smaller for the nanotubes with smaller radii.

We expect to continue investigation of magnetic properties of the different forms of the nanostructures in the near future.

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