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Theory of weakly coupled two-dimensional magnets

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Abstract

We demonstrate that the experimental findings of the magnetic properties of the weakly coupled trilayer system $Ni_4/Cu_N/Co_2$ are reproduced by a theory that combines first principles calculations of the exchange interactions in a classical Heisenberg model with Monte Carlo simulations. Through an analysis of the spin–spin correlation function we show that two distinct temperatures can be identified; a higher temperature where long range magnetic order disappears and a lower temperature where the spin–spin correlation of the Ni atoms undergoes a drastic change. We argue that our findings hold in general for 'weak exchange link' systems.

1. Introduction

The study of magnetism in reduced dimensions has attracted a lot of attention during past decades [1], not least due to the ability to prepare thin film materials in a well controlled way and to detect the magnetic properties in an element-specific way, as, for example, is done by magnetic circular x-ray dichroism (MCXD). These experimental results can be compared to theoretical models, in particular how the strong influence of the reduced dimensionality of thin films (that are essentially two-dimensional materials) modifies the magnetic ordering [2].

Recent experimental studies of weakly coupled two-dimensional magnets [3–7] in combination with theoretical analysis [8, 9] have added to our knowledge of these systems. The experimental work consisted of the growth of thin layers of Co and Ni sandwiched with Cu layers of varying thickness, which acted as a weak exchange link between the Co and Ni atoms. These trilayers were grown in an fcc crystal structure on top of a Cu(001) substrate. By means of an element-specific probe (MCXD) the magnetic moments of both the Ni layer and the Co layers could be followed, both as a function of temperature and as a function of coupling strength (via the interlayer exchange coupling) between the Co and Ni layers. In [3] and [4] it was found that the magnetism of the Ni layers disappeared at a lower temperature than that of the Co layers. This led the authors of [3] and [4] to define a temperature T^* where the Ni moment disappeared, which is substantially lower than the critical (Curie) temperature, T_C, of the trilayer. It is of-course tempting to assign two different critical temperatures to this system, one for Ni atoms and one for Co atoms, especially since the magnetization curve of Ni up to

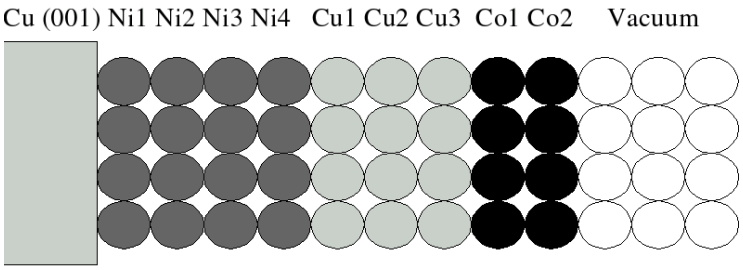

Figure 1. Geometry of the calculations of the trilayer system, in this case $Ni_4/Cu_3/Co_2$ on top of a Cu(001) substrate. The labelling of the atomic layers is found at the top of the figure (Ni1, Ni2, ...).

T^* was found to be very similar to magnetization curves in general of thin films close to the ordering temperature. However, using thermodynamic arguments only one critical temperature of the trilayer was identified, with a stipulated definition of a second temperature called T^* when the Ni moments appeared to become disordered.

The present paper provides a microscopic analysis of the experimental works of [3–7] using a combination of first principles theory, a classical Heisenberg Hamiltonian and Monte Carlo simulations. In contrast to previous theory [8, 9] we make no assumptions about the exchange interactions in these systems, instead we calculate from first principles all the properties required (magnetic moments and exchange interactions) for our finite temperature analysis. As mentioned, a theoretical analysis for the experimental works in [3–7] has been provided in [8, 9]. Although we certainly do not claim that these theoretical works are erroneous in any way, we nevertheless feel that we have added substantial and new information. As we shall see below, we come to the conclusion that theory reproduces on a qualitative level the observations in [3–7] and that the spin–spin correlation vanishes in the Ni layers in the temperature interval between T^* and T_C, so that an assignment of one critical temperature for the Ni layer and another critical temperature for the Co layer is a rather practical way to analyse the results of [3–7] and of similar systems.

Since the present study concerns the calculation of the critical temperature of layered materials we draw the reader's attention to the fact that methods based on the molecular field approximation have been used previously to address such issues. The Curie temperatures of compositionally modulated multilayers were found to depend strongly on film thickness, modulation and interdiffusion [10–12]. Also, the theoretical work in [8, 9] was based on the Tyablikov approximation. Finally we note that the approach taken here has previously been used for Fe–V multilayers [13].

2. Theory

The considered geometry of the calculations is shown in figure 1. It is well established that the atomic species of these thin film systems are situated on an fcc lattice. Structural relaxations are small and have for simplicity been ignored in this work. We have considered geometries composed of four layers of Ni on a Cu(001) substrate, separated by n layers of fcc Cu (where $n = 1, 2$ and 3) followed by two layers of fcc Co at the surface of the samples. This is exactly the geometry considered experimentally in [3]. The first principles calculations make use of the all-electron Korringa–Kohn–Rostoker (KKR) Green's function method in combination with the multipole corrected atomic sphere approximation (ASA) [14]. The generalized gradient

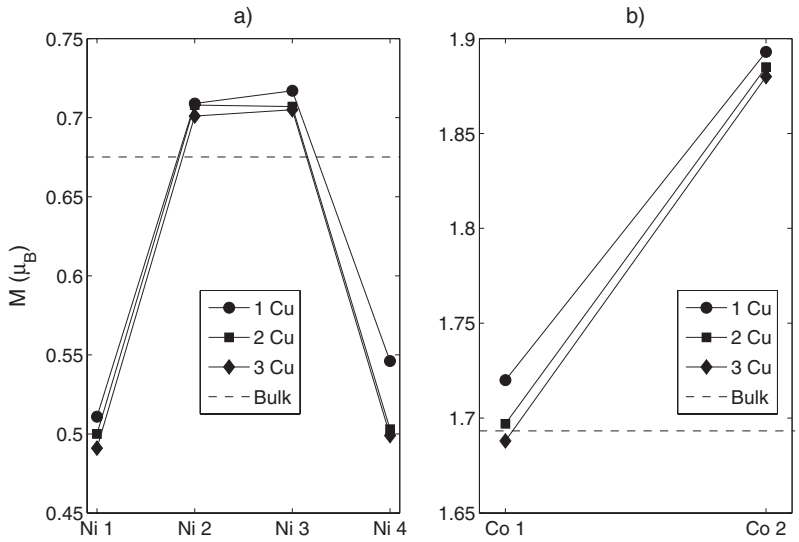

Figure 2. Calculated layer resolved magnetic moments in Bohr magnetons of the (a) Ni and (b) Co layers in the trilayer system for different numbers of Cu spacer layers. The curves denoted n Cu ($n = 1, 2$ or 3) correspond to a trilayer where the Ni and Co atoms are separated with n Cu layers. The same notation as in figure 1 for the different layers is used. The calculated bulk magnetic moments are displayed by a dotted line.

approximation as parameterized by Perdew, Bunke and Enzerhof (PBE96) was employed [15] for the exchange correlation potential. The valence basis set consists of s, p and d orbitals where scalar relativistic effects are taken into account but neglecting spin orbit effects. In a self-consistent manner both the magnetic moments, the intralayer (interatomic) exchange energies and the interlayer exchange energies have been calculated. The exchange interactions in the Heisenberg model, J_{ij}, were calculated directly using the magnetic force theorem and the Green's function employing the equation by Lichtenstein *et al* [17]. The intralayer exchange interaction has also been calculated from the difference in total energy between the ferromagnetic and antiferromagnetic configuration of the Ni and Co layers, respectively.

3. Results

The self-consistently calculated magnetic moments are displayed in figure 2, and are found to agree well with previous theory [16] and with experiment [3–7]. The calculated exchange parameters and magnetic moments were then used in a classical Heisenberg Hamiltonian, for which the thermodynamic problem was solved by means of Monte Carlo simulations [18] (with periodic boundary conditions in the film plane and free boundary conditions in the z-direction). It should be noted that the mean field theory is not appropriate for low-dimensional systems and a theory that properly includes fluctuations is needed, like the Monte Carlo simulations.

In figure 3 we show, as an example, the calculated exchange parameters of the $Co_2Cu_3Ni_4$ system as a function of distance. In figure 3(a) we show the exchange parameters within each of the four Ni layers. Note that the atoms labelled Ni2 and Ni3 only have Ni atoms as nearest neighbours whereas the atoms labelled Ni1 and Ni4 also have Cu atoms as nearest neighbours. For this reason the exchange parameters of the Ni2 and Ni3 atoms are similar and the parameters of Ni1 and Ni4 atoms are similar. In figure 3(b) the exchange parameters

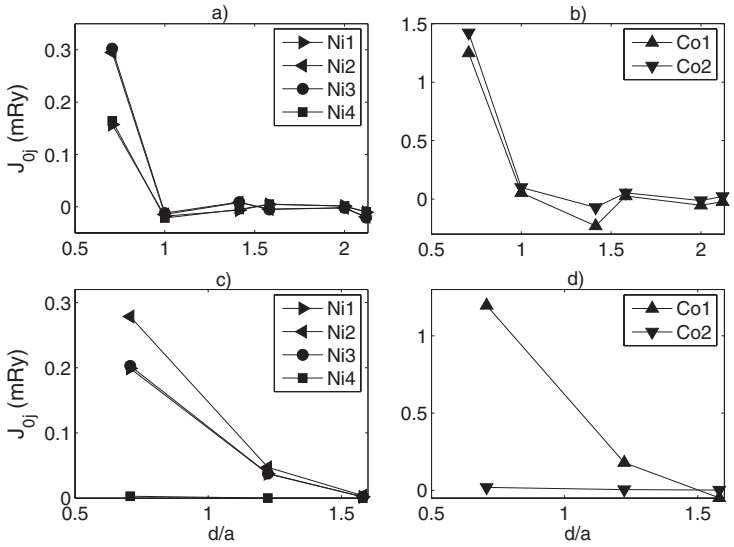

Figure 3. Calculated exchange parameters for the $Co_2Cu_3Ni_4$ system as a function of interatomic distance (d) normalized with the lattice constant (a). For details see text.

within the two Co layers are shown. In figure 3(c) we display the exchange parameters between atoms within two neighbouring Ni layers as a function of distance between these atoms. The data labelled Ni1 show the exchange parameters between atoms in the Ni1 layer and atoms in the Ni2 layer. In a similar way the data labelled Ni2 depict the exchange parameter between atoms in the Ni2 layer and atoms in the Ni3 layer. Since the data points labelled Ni4 refer to the coupling between the Ni4 atoms and an essentially non-magnetic Cu, this exchange is nearly zero. In a similar way the data in figure 3(d) show the exchange interaction between two neighbouring Co layers, and the data labelled Co1 refer to the exchange between atoms in the Co1 layer and the atoms in the Co2 layer. All data in figure 3 show that the nearest neighbour exchange is dominating but that the longer ranged interactions are also important. We have chosen not to show similar data for the $Co_2Cu_2Ni_4$ system and the $Co_2Cu_1Ni_4$ system, since the data are similar to those in figure 3. Finally we note that the interlayer exchange interaction is 0.1 mRyd. All exchange interactions up to two lattice constants ($d/a = 2$) were included in the Monte Carlo simulations.

Before we describe the results for the systems with a weak exchange link, we tested our method by comparing calculated critical temperatures of thin films of fcc Ni on Cu(001) that were capped with two overlayers of Cu. Experimentally it has been established that four layers of Ni with two overlayers of Cu on top of a Cu(001) substrate result in a critical temperature of around 200 K [3], which is reduced compared to the bulk value (634 K). Our theoretical value for the same system is 209 K, deduced from finite-size scaling using the cumulant-crossing method [18], which is in very good agreement with experimental data. The agreement is somewhat fortuitous since the present calculation scheme, using exchange parameters from the equation by Lichtenstein and the gradient generalized approximation in combination with Monte Carlo simulations, underestimates T_C of bulk Ni by a fair amount (400 K compared to 634 K experimentally).

If on top of these Ni and Cu layers one deposits two layers of fcc Co, the temperature for when the magnetism of the Ni layers disappears is increased to around 260 K [3], which should

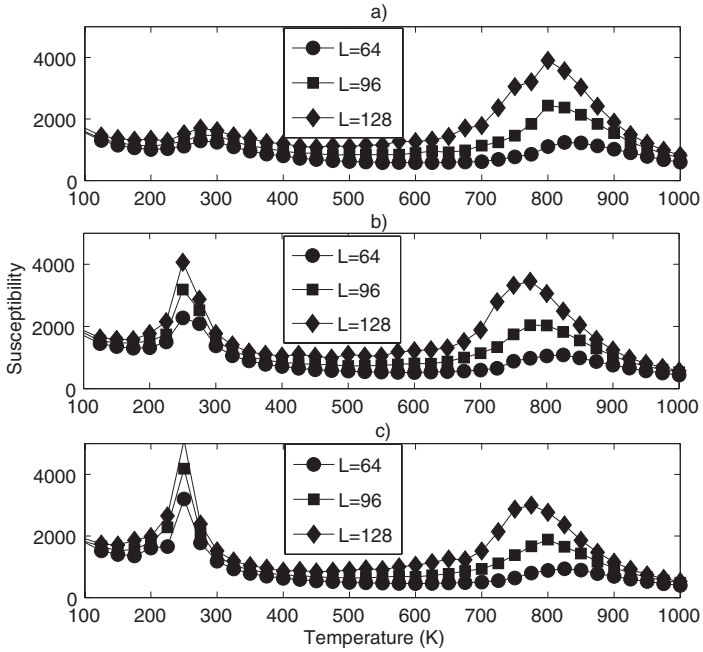

Figure 4. Susceptibility of the $Co_2/Cu_n/Ni_4$ trilayer system for (a) $n = 1$, (b) $n = 2$ and (c) $n = 3$, plotted for three different lattice sizes (number of spins in plane is $L \times L$).

be compared to our theoretical values of around 250 K. In other words, we can conclude that the exchange field produced by the two layers of Co increases the ordering temperature (or T^*) by around 60 K in the experimental study and by 40 K in our calculations.

In figure 4 we display the calculated magnetic susceptibility χ, defined as $\chi = \frac{N}{k_B T}(\langle M^2\rangle - \langle|M|\rangle^2)$, where N is the total number of spins in the lattice, T is temperature and M is magnetization, for the three weak exchange link systems ($Co_2Cu_nNi_4$, $n = 1$–3). Two conspicuous peaks in the susceptibility may be seen from figure 4, one at ∼250 K and one at ∼750 K (the position of the peaks for different lattice sizes shifts slightly due to finite size effects). This indicates two magnetic transitions. It may also be noted from figure 4 that in the system with strongest interlayer exchange coupling ($n = 1$) the peak at ∼250 K is rather broad. This is in contrast to the $n = 3$ case where the peak at ∼250 K is sharp and can better be distinguished from the ∼750 K peak.

In order to analyse on a microscopic level the origin of the two peaks in figure 4 we show in figure 5 the in-plane spin–spin correlation function ($\langle s_i \cdot s_j\rangle$, where i and j are the atomic indices). In the figure we have evaluated the spin–spin correlation function for a $Co_2/Cu_3/Ni_4$ trilayer at two temperatures (at 225 and 700 K), the first just below the first peak in the susceptibility (the 250 K peak) and the second just below the second peak (the 750 K peak). It should be noted that the spin–spin correlation is calculated for all four Ni layers and for both Co layers, producing the six curves that are shown in figure 5. The figure shows that at 225 K there is significant spin–spin correlation in both the Ni and Co layers. At 700 K this correlation is more or less absent for the Ni layers, whereas for the Co layers it is still quite noticeable. Hence it is clear that the peak at low temperatures in figure 4 is associated with the disappearance of magnetism in the Ni layers, whereas the second peak (∼750 K) is associated with the disappearance of the Co magnetism. Since the spin–spin correlation for the Ni layers

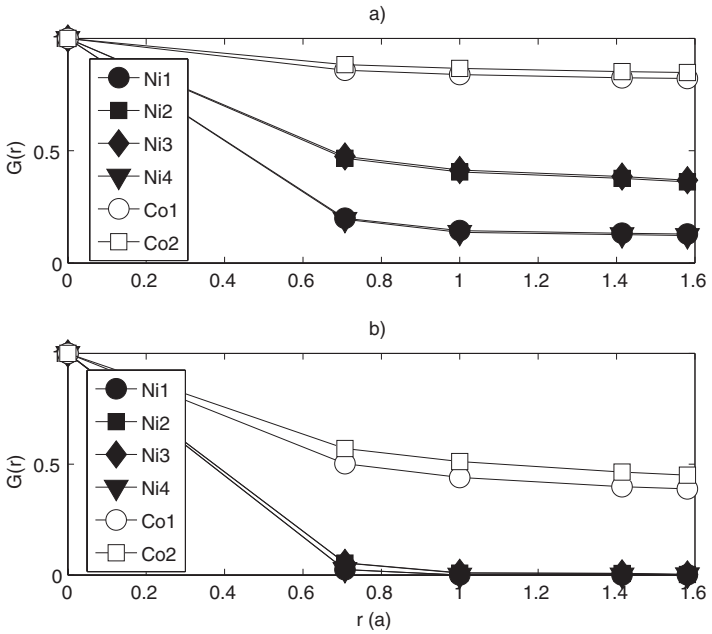

Figure 5. The spin–spin pair correlation function in-plane plotted as a function of distance (measured in lattice constants) of the $Ni_4/Cu_3/Co_2$ trilayer system for (a) $T = 225$ K and (b) $T = 700$ K. The notation of the different layers is the same as in figure 1.

at 700 K is more or less identical to that of a four layer Ni film (without interaction from Co layers) above the ordering temperature (data not shown), it is tempting to conclude that the first peak in the susceptibility in figure 4 represents the ordering temperature of the Ni layers whereas the second peak corresponds to the ordering temperature of the Co layers.

In order to further analyse the situation we show in figure 6 the spin–spin correlation for the $Co_2/Cu_3/Ni_4$ trilayer in the z-direction, i.e. along the direction between the Ni and Co layers. In this figure we have positioned the atom of interest at the origin in the plot. This means that the spin–spin correlation between, for example, the surface Co atom (Co2, given by open squares) and the other atoms (Co and Ni) is plotted at negative distances. In contrast, the correlation function between the subsurface Co atom (Co1, given by open circles) has the correlation function with the Co2 atom at positive z-values, whereas the correlations with the Ni (and Cu) atoms are located at negative z-values. In a similar fashion the Ni1 atom has non-zero spin–spin correlations only for positive z-values, since only these values correspond to other Ni or Co atoms (at negative z-values there are only Cu atoms, with an almost vanishing magnetic moment). A closer inspection of figure 6 shows that at 225 K there is a non-vanishing coupling between the atoms of the Ni and Co layers. This can be seen in the Co1 correlation function, since the data point at -2 on the x-axis represents the correlation between the interface Co and interface Ni atoms over the Cu layers (the same information is given by the Ni4 correlation function). At 700 K the situation is different, there are still correlations within the Co layers, but between the Co and the Ni layers there is a vanishingly small spin–spin correlation.

4. Summary

In summary we have provided a detailed microscopic analysis of thin film materials with a ‘weak exchange link’. From first principles theory we have evaluated all relevant, material-

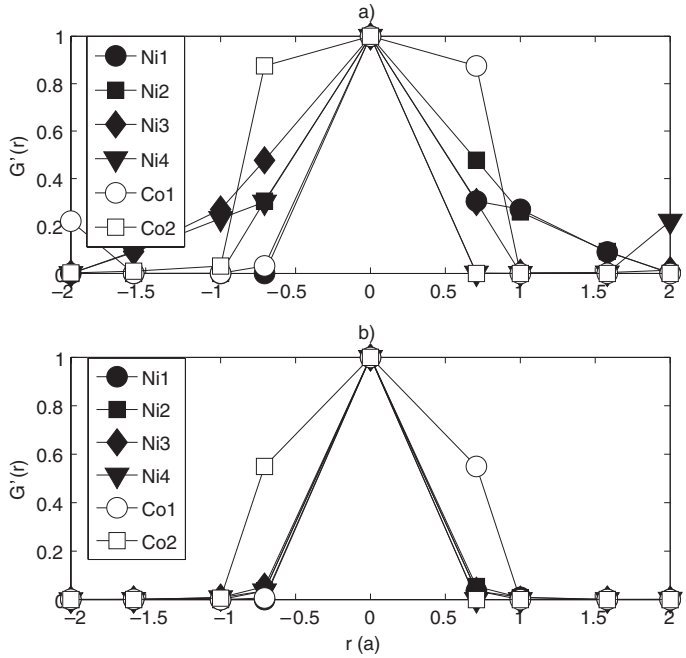

Figure 6. Spin–spin pair correlation function in the z-direction as a function of distance (measured in lattice constants) of the $Ni_4/Cu_3/Co_2$ trilayer system for (a) $T = 225$ K and (b) $T = 700$ K. The notation of the atoms in the different layers is the same as in figure 1.

specific information for use in a classical Heisenberg model and we have reproduced on a quantitative level the magnetic properties (spin moments and ordering temperatures) of these systems. Via an analysis of the spin–spin correlation we come to the conclusion that two distinct temperatures are relevant to describe these systems, one representing the critical temperature of the entire system and a lower temperature where the spin–spin correlation for a smaller part of the system (the Ni atoms) undergoes an abrupt modification. Although we have focused on the experimental studies of Co/Cu/Ni systems we believe that our analysis most likely also holds for other 'weak link' systems.

At first glance our results are at variance with the analysis of nanocomposites presented by Skomski and Sellmyer [19], where only one critical temperature was identified in a system containing two atomic species. In their study a composite material, containing certain atoms with strong exchange interaction and certain atoms with weak exchange interaction, was analysed in a mean field approximation. It was shown that this model results only in one common critical temperature, with no sign of a second distinct temperature, T^*. However, this study was focused on magnetic atoms in direct contact, which is not quite the situation discussed in the present work, since we are considering magnetic Ni and Co atoms separated by non-magnetic Cu layers. Hence, apart from having a more precise theoretical treatment, we are here focusing on a system where the exchange field between the two different magnetic species (Ni and Co) is given by the interlayer exchange interaction [20, 21], and it is hence much weaker than the exchange field calculated in [19].

The present work also shows that the ordering temperatures of thin film and surface systems is accessible from first principles theory, in a similar way to what is known for bulk [22] and multilayer [13] systems. Finally we note that since the critical temperature of Ni can be

made to be larger than that of the Co layers, if one increases the Ni thickness, and since the Ni moments are smaller than the Co moments, one could envisage a situation where for a certain temperature the moment of the Ni layers is identical to that of the Co layers. If then the spacer (Cu) thickness is tuned so that the Ni and the Co layers couple antiferromagnetically one could identify a temperature where the total moment of the film is zero. In other words one could identify a compensation temperature, where the film would possibly exhibit a diverging coercive field, such as is found for certain rare-earth transition metal systems [23]. A realization of this situation may be important for magnetic recording.

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