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Theoretical approach to the Curie temperature shift in FM1/NM/FM2/SUB (ferromagnetic metal 1/nonmagnetic metal/ferromagnetic metal 2/substrate) systems

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Abstract. An itinerant-electron model is proposed for investigating the Curie temperature shift in indirectly coupled itinerant ferromagnetic metal/nonmagnetic metal/ferromagnetic metal (FM2)/substrate structures. The Coulomb correlation between the electrons in the ferromagnetic metal is treated by using the spectral density approach. The magnetic susceptibility is used to determine the Curie temperature shift due to the interlayer exchange coupling. The relation between the Curie temperature shift and the interlayer exchange coupling is studied. It shows that the Curie temperature shift is related to the strength of the interlayer exchange coupling due to FM2 sublayers other than that of the whole system at high temperature. Good agreements with experiments are obtained.

1. Introduction

The magnetic properties of metallic magnetic multilayers have given rise to a lot of studies. Most of these studies focus on the interlayer exchange coupling [1, 2] (IEC), giant magnetoresistance [3,4] (GMR), and quantum well effect [5–8]. There has been little work on the effect of IEC on the magnetic properties of ferromagnetic sublayers (FM). However, recently, it was found that the magnetic properties of FMs were changed due to the IEC—the Curie temperature of the FM had a shift from its uncoupled value [9, 10]. In these experiments, two FMs were separated by a nonmagnetic spacer (NM) mediating the IEC. The magnetic properties of these two FMs are different in the uncoupled cases—e.g., the magnetizations ($m_{FM1} \neq m_{FM2}$) and Curie temperatures ($T_{C1} > T_{C2}$). From the curve of magnetization as a function of temperature, two Curie temperatures (T^*_{C1} and T^*_{C2}) were observed in FM1/NM/FM2/SUB (where SUB refers to the substrate sublayers) systems. The magnetization of FM2 goes to zero at $T = T^*_{C2}$ with $T^*_{C2} > T_{C2}$. The difference $\Delta T_{C2} = T^*_{C2} - T_{C2}$ is defined as the shift of the Curie temperature of FM2. The magnetic susceptibility as a function of temperature $\chi(T)$ is also measured in the above experiments [9, 10]. In the weakly coupled case (large thickness of the NM), the magnetic susceptibility displays two maxima: one singularity at T^*_{C1} near T_{C1}; one maximum at T^*_{C2} near T_{C2}. However, in the strongly coupled case only one singularity in the curve of $\chi(T)$ was measured at T^*_{C1}. Now there exists a contradiction in the above results. If there exist two Curie temperatures, $\chi(T)$ must display two singularities at different

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temperatures. On the other hand, if there exists only one Curie temperature, the magnetization of FM2 should not go to zero at $T = T^*_{C2}$.

For the directly coupled FM1/FM2 superlattice [11], the temperature dependence of the thermal expansion coefficient $\alpha_{[001]}(T)$ was measured. The data display either two anomalies or one anomaly depending on the composition d_{FM1}/d_{FM2} (d_{FM1} and d_{FM2} are the numbers of layers of FM1 and FM2, respectively). In order to interpret the experimental results for the directly coupled systems, the localized spin model was treated in the mean-field approximation [12, 13]. Within Ginzburg–Landau theory, Wang and Mills [12] studied the onset of long-range order in a directly coupled superlattice. In some cases, $\chi(T)$ has two maxima, one at T^*_{C1}, the other at T^*_{C2}. But only one singularity was obtained in $\chi(T)$. The magnetizations of these two FMs are nonzero in the temperature range between these two $\chi(T)$ maxima. The structure can exhibit only one true Curie temperature T^*_{C1}, and below this temperature long-range order exists everywhere in the FM1/FM2 superlattice.

We have mentioned that the above theories [12, 13] are based on the localized spin models and use the mean-field approximation. However, in transition metal multilayers the magnetically active electrons are itinerant. It is by no means clear to what extent the results obtained from localized spin models are applicable to transition metal films. Furthermore, in the theory of IEC [14, 15], only the properties of NM are considered, while the discrete magnetic properties of FMs are completely neglected.

In this paper, we study the change of magnetic properties of FMs due to the IEC within the single-band Hubbard model [16]. Since a simple Hartree–Fock (Stoner) theory is known to overestimate the possibility of ferromagnetic order drastically and even to lead to qualitatively wrong results [17], a more sophisticated treatment of the Coulomb correlation between electrons in the FMs is needed. Here we employ the so-called spectral density approach (SDA) [18, 19]. The SDA leads to qualitatively reasonable results for the magnetic properties of the Hubbard model [18–23]. A detailed study of ferromagnetism within the SDA and a comparison with the results from different analytical approximation schemes as well as with numerical exact quantum Monto Carlo (QMC) results has been performed for the Hubbard model on infinite-dimensional lattices [24]. It has been found that for a reasonable description of finite-temperature ferromagnetism it is vital to be consistent with the perturbation theory of Harris and Lange [25] which provides exact results for the strong-coupling limit. Crucial for ferromagnetic order is the possibility for a spin-dependent shift of the centres of gravity of the Hubbard bands. This 'band shift' is neglected within most analytical approaches but correctly included within the SDA [24].

We expect this to be the main mechanism for ferromagnetic order also in the case of finite-dimensional lattices and for three-dimensional thin films in particular. While the SDA considerably improves upon a simple Hartree–Fock treatment, it still has to be classified as a mean-field-type approximation scheme. Like any mean-field approach, however, it is therefore conflicting with the theorem of Mermin and Wagner [26] which excludes finite-temperature ferromagnetism for the isotropic Hubbard model in multilayers of finite thickness [27]. On the other hand, for real materials the theorem is quite irrelevant since (small but) finite anisotropic interactions are always present. The so-called reorientation transition in the Hubbard model with additional dipole and spin–orbit interaction has been studied by means of the SDA recently [20]. As long as one is not interested in the direction of the magnetization, however, it is not necessary to include anisotropic interactions. The global spin-rotation symmetry is broken by the mean-field (SDA) approximation itself. Of course, this implies that one can expect qualitatively reasonable results at best, and that a detailed agreement with experimental results cannot be expected.

The paper is organized as follows. First, the Hamiltonian of our model is proposed, and

the SDA for the Hubbard film is described in a simple way. In section 3 we show the results of the numerical evaluation of the theory. Finally, a summary will be given.

2. Theoretical model

The theoretical method used is similar to that described previously [19–23], so we only give a brief derivation of our theory.

In the film system, each lattice vector is decomposed into two parts:

$$\boldsymbol{R}_{i\alpha} = \boldsymbol{R}_i + \boldsymbol{r}_\alpha \tag{1}$$

where $\boldsymbol{R}_i$ denotes a lattice vector of the two-dimensional Bravais lattice of the surface layer with N sites and $\boldsymbol{r}_\alpha$ the vector of the centre of the αth layer. Within each layer we assume translational invariance.

Within the Hubbard model, and considering the difference of the FMs and NM, we choose the Hamiltonian as follows:

$$\mathcal{H} = \sum_{i,j,\alpha,\beta,\sigma} (T_{ij}^{\alpha\beta} - \mu\delta_{ij}^{\alpha\beta}) c_{i\alpha\sigma}^{+} c_{j\beta\sigma} + \frac{1}{2} \sum_{i,\alpha,\sigma} U(\alpha) n_{i\alpha\sigma} n_{i\alpha-\sigma} \tag{2}$$

where $c_{i\alpha\sigma}^{+}$ ($c_{i\alpha\sigma}$) stands for the creation (annihilation) operator for an electron with spin σ at the lattice site $\boldsymbol{R}_{i\alpha}$, $n_{i\alpha\sigma} = c_{i\alpha\sigma}^{+} c_{i\alpha\sigma}$ is the number operator, and $T_{ij}^{\alpha\beta}$ denotes the hopping-matrix element connecting the lattice sites $\boldsymbol{R}_{i\alpha}$ and $\boldsymbol{R}_{j\beta}$. μ is the chemical potential.

Here the on-site Coulomb interaction $U(\alpha)$ is layer dependent; the dependence is different for the FMs and NM:

$$U(\alpha) = \begin{cases} 0 & \alpha \in \text{SUB or NM} \\ U_1 & \alpha \in \text{FM1} \\ U_2 & \alpha \in \text{FM2.} \end{cases} \tag{3}$$

The basic quantity to be calculated is the retarded single-electron Green function

$$G_{ij\sigma}^{\alpha\beta}(E) = \langle\langle c_{i\alpha\sigma}; c_{j\beta\sigma}^{+} \rangle\rangle_E \tag{4}$$

which includes all relevant information about the system.

The equation of motion for the single-electron Green function reads

$$\sum_{l,\gamma} [(E+\mu)\delta_{il}^{\alpha\gamma} - T_{il}^{\alpha\gamma} - \Sigma_{il\sigma}^{\alpha\gamma}(E)] G_{lj\sigma}^{\gamma\beta}(E) = \hbar\delta_{ij}^{\alpha\beta}. \tag{5}$$

Here we have introduced the electronic self-energy $\Sigma_{ij\sigma}^{\alpha\beta}(E)$ which incorporates all effects of electron correlations.

The key point of the SDA is to find a reasonable *ansatz* for the self-energy in FMs. Guided by the exactly solvable atomic limit of vanishing hopping ($t_{ij}^{\alpha\beta} = 0$) and by the findings of Harris and Lange [25] in the strong-coupling limit, a one-pole *ansatz* for the self-energy $\Sigma_\sigma^\alpha(E)$ can be motivated [22]:

$$\Sigma_\sigma^\alpha(E) = g_{1\sigma}^\alpha \frac{E - g_{2\sigma}^\alpha}{E - g_{3\sigma}^\alpha} \tag{6}$$

where the spin- and layer-dependent parameters $g_{1\sigma}^\alpha$, $g_{2\sigma}^\alpha$, and $g_{3\sigma}^\alpha$ are fixed by exploiting the equality between two alternative but exact representations for the moments of the layer-dependent quasi-particle density of states (QDOS). It has been shown [24] that inclusion of the first four moments of the QDOS ($m = 0$–3) is vital for a proper description of ferromagnetism in the Hubbard model, especially for finite temperatures. Further, the first four moments represent a necessary condition [24] for consistency with the strong-coupling results of Harris

and Lange. Taking into account the first four moments to fix the three parameters in (6), one obtains the SDA self-energy [22–24], which depends on the spin-dependent occupation numbers $n_{\alpha\sigma}$ and the so-called band shift $B_{\alpha\sigma}$ that consists of higher correlation functions.

The problem is that of how to find a self-consistent set of $n_{\alpha\sigma}$ and $B_{\alpha\sigma}$. Ferromagnetism is indicated by a spin asymmetry in the band occupations $n_{\alpha\sigma}$ leading to nonzero layer magnetizations $m_\alpha = n_{\alpha\uparrow} - n_{\alpha\downarrow}$. The mean magnetization of FM1 is defined as

$$m_{FM1} = \sum_{\alpha \in FM1} m_\alpha / d_{FM1}.$$

The same definitions are used for FM2 and NM. The band occupation in each layer is given by $n_\alpha = n_{\alpha\uparrow} + n_{\alpha\downarrow}$.

The internal energy E:

$$E = \frac{\langle \mathcal{H} \rangle}{Nd} = \frac{1}{Nd}\left[\sum_{i,\alpha,j,\beta,\sigma} (T_{ij}^{\alpha\beta} - \mu\delta_{ij}^{\alpha\beta})\langle c_{i\alpha\sigma}^{+} c_{j\beta\sigma}\rangle + \frac{1}{2}\sum_{i,\alpha,\sigma} U(\alpha)\langle n_{i\alpha\sigma} n_{i\alpha-\sigma}\rangle\right] \tag{7}$$

is used to define the interlayer exchange coupling. The above expression can be obtained via spectral theorem from the one-electron Green function:

$$E = -\frac{1}{\pi d\hbar}\,\mathrm{Im}\sum_{\alpha,\sigma}\int_{-\infty}^{\infty} \mathrm{d}E\; f_{-}(E)\left[\left(E - \mu - \frac{1}{2}\Sigma_\sigma^\alpha(E-\mu)\right)G_{ii\sigma}^{\alpha\alpha}(E-\mu) - \hbar\right]. \tag{8}$$

3. Results and discussion

The systems investigated are: FM1/NM/FM2/SUB (S1), FM1/NM/NM2/SUB (S2), FM1/NM (S3), and NM/FM2/SUB (S4) (see figure 1). We will refer to the above systems as S1: $d_{FM1}/d_{NM}/d_{FM2}/d_{SUB}$, S2: $d_{FM1}/d_{NM}/d_{NM2}^{r}/d_{SUB}$, S3: d_{FM1}/d_{NM}, and S4: $d_{NM}/d_{FM2}/d_{SUB}$, respectively. In S2, the parameters used for NM2 are the same as those used for FM2 in S1 except for the Coulomb interaction. An fcc(100) geometry is assumed for the whole systems. The parameters used in all figures are as follows: $U_1 = 4.0$ and $U_2 = 6.0$ which are taken near the values for Co and Ni [28]. $n_{FM1} = 1.5$ and $n_{FM2} = 1.8$ can lead to similar magnitudes of T_C to those found for Co and Ni, respectively. $n_{NM} = 1.5$ and $t = 0.1$ eV. Different values of the parameters were also treated in our calculations, yielding qualitatively the same results.

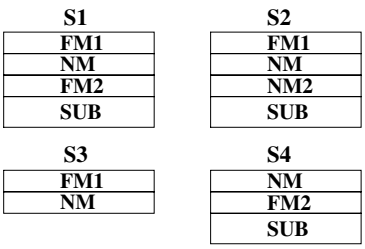

Figure 1. The systems investigated, shown schematically, namely: S1: FM1/NM/FM2/SUB; S2: FM1/NM/NM2/SUB; S3: FM1/NM; and S4: NM/FM2/SUB. The properties of NM2 in S2 are the same as those of FM2 in S1 except for the Coulomb interaction—$U_{NM2} = 0$.

First, let us discuss the temperature dependence of the mean magnetizations of FM1 and FM2 for S1 and S4. In figure 2(a), we plot the mean magnetizations of FM1 and FM2 as functions of temperature ($m_{FM1}(T)$ and $m_{FM2}(T)$) for an uncoupled system (S4) and a coupled system (S1). $m_{FM2}(T)$ goes to zero at T_{C2} in S4 (dashed line). The system displays a

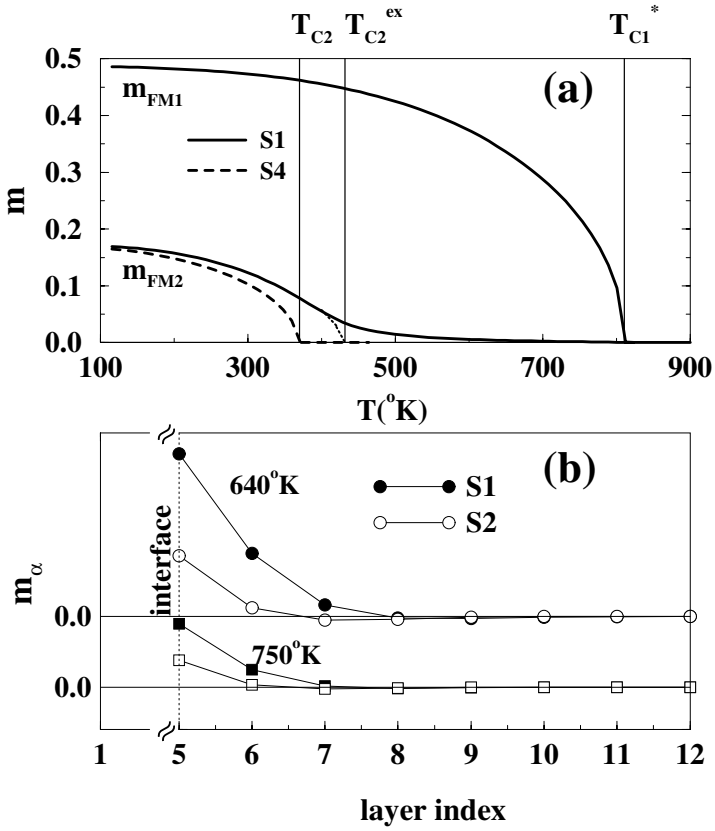

Figure 2. (a) The mean magnetizations of FM1 and FM2 as functions of temperature for S1: 2/2/4/6 and S4: 2/4/6 and (b) the magnetization profile of FM2 and NM2 at $T = 640$ K and $T = 750$ K for S1: 2/2/8/6 (filled symbols) and S2: 2/2/8^r/6 (open symbols). The dotted line in (a) is an extrapolated behaviour of m_{FM2} which corresponds to the experimental results of reference [9].

true transition temperature T_{C2}, while in S1, $m_{FM2}(T)$ does not go to zero until the temperature reaches T^*_{C1} (solid line). At T_{C2}, $m_{FM2}(T)$ is large due to the strong IEC between FM1 and FM2. Above T_{C2}, $m_{FM2}(T)$ drops gradually, not, however, becoming exactly zero. We note that the indirectly coupled system can exhibit only one Curie temperature T^*_{C1}. Below T^*_{C1}, the long-range order exists everywhere in the whole system. So we cannot define T^*_{C2} from the curve of $m_{FM2}(T)$ as can be done in experiments [9, 10]. According to the extrapolation of $m_{FM2}(T)$ above T_{C2}, we obtain a dotted line m^{ex}_{FM2} in figure 2(a) which corresponds rather well to experimental results [9]. One obtains a temperature T^{ex}_{C2} at the zero point of m^{ex}_{FM2}. But we will see later that this temperature is larger than the temperature obtained from the maximum of the magnetic susceptibility.

It is interesting to study the long-range order in FM2 at temperatures $T_{C2} \ll T < T^*_{C1}$, because in this temperature range, the thermal fluctuation should have destroyed the long-range order of FM2. The long-range order can only be induced by the IEC. However, it is an interesting question whether or not the Coulomb interaction within FM2 will still affect the magnetization of FM2. In order to answer this question, we calculated the magnetization profile of S1: 2/2/8/6 and S2: 2/2/8^r/6 at $T_{C2} \ll T < T^*_{C1}$ (see figure 2(b)). Comparing these two cases, the magnetizations of FM2 and NM2 are quite different for several layers close to

the interface. This means that the Coulomb correlation within FM2 still plays an important role for the magnetic properties of FM2 in this temperature range. As temperature increases, the effect of Coulomb interaction on the magnetic properties of FM2 becomes weaker (see figure 2(b)).

In order to investigate the response of the indirectly coupled system, we calculated the magnetic susceptibility as a function of temperature $\chi(T)$. The magnetic susceptibility can be obtained by adding a Zeeman term in the Hamiltonian. In figure 3, $\chi(T)$ and $1/\chi(T)$ are plotted for different numbers of NM layers in S1: $2/d_{NM}/4/6$. There are two maxima in the $\chi(T)$; one is a singularity which corresponds to the real Curie temperature, the other is located at T^*_{C2} near to T_{C2}. Even in the strongly coupled cases ($d_{NM} < 3$), $\chi(T)$ still displays two maxima. However, since the magnitude of the maximum (χ_{max}) at T^*_{C2} is very small, experimentally it will be confused with the background. But it can be seen clearly in $1/\chi(T)$. When the number of NM layers increases, χ_{max} at T^*_{C2} tends to increase with a little oscillation. Its relation to the IEC will be discussed below. Defining the temperature T^*_{C2} as a quasi-Curie

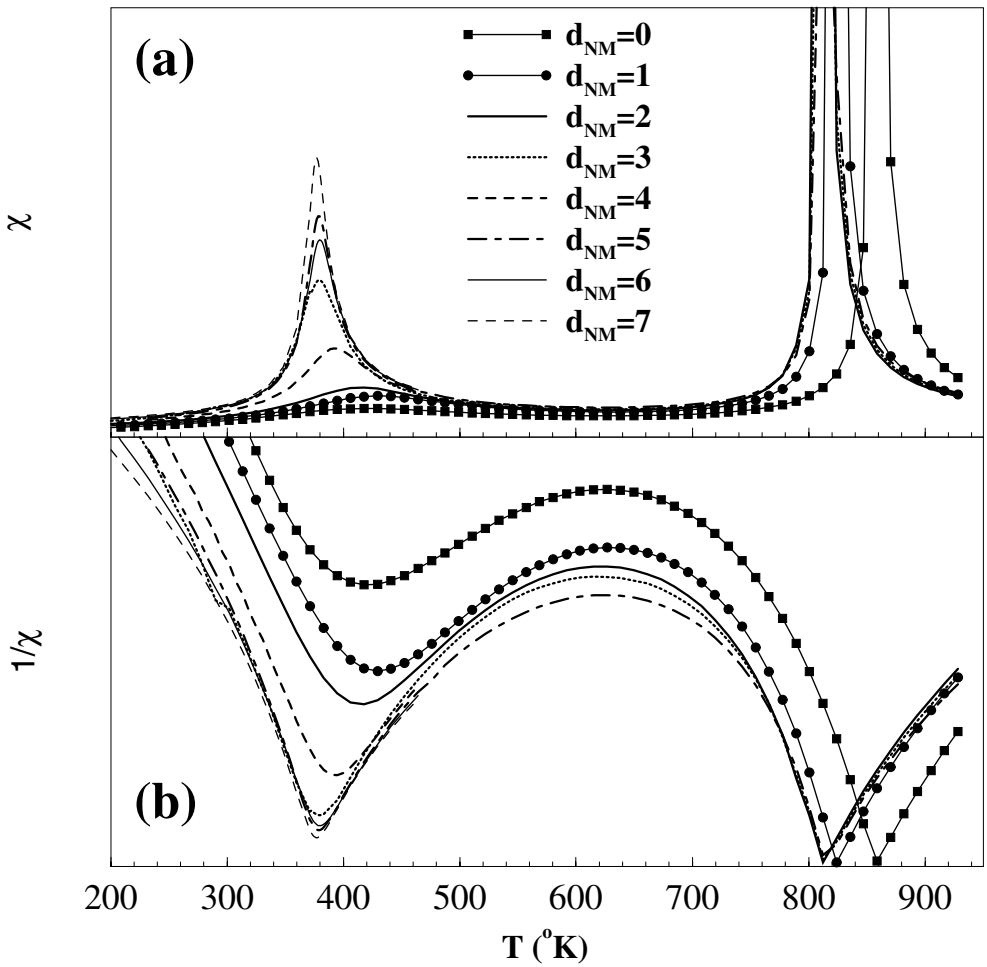

Figure 3. The magnetic susceptibility $\chi(T)$ (a) and its inverse (b) as functions of temperature for S1: $2/d_{NM}/4/6$ ($d_{NM} = 0, \ldots, 7$).

temperature†, and $\Delta T_{C2} = T^*_{C2} - T_{C2}$ as the Curie temperature shift, we can find that ΔT_{C2} relates to χ_{max} at T^*_{C2}. The larger χ_{max} is, the smaller ΔT_{C2} is, and in the case of a large number of NM layers, $\Delta T_{C2}\,\chi \approx$ constant (see figure 4(c)). At high temperature, the Curie temperature T^*_{C1} of the coupled case (S1) is different from T_{C1} for the uncoupled case (S3). Here we would like to point out that we cannot use the bulk Curie temperature T^B_{C1} instead of T_{C1} in discussing the influence of the IEC on the FM1 [12]. This is because the Curie temperature for films is quite different from that for the bulk case. Further, we must include the influence of the NM on the FM1.

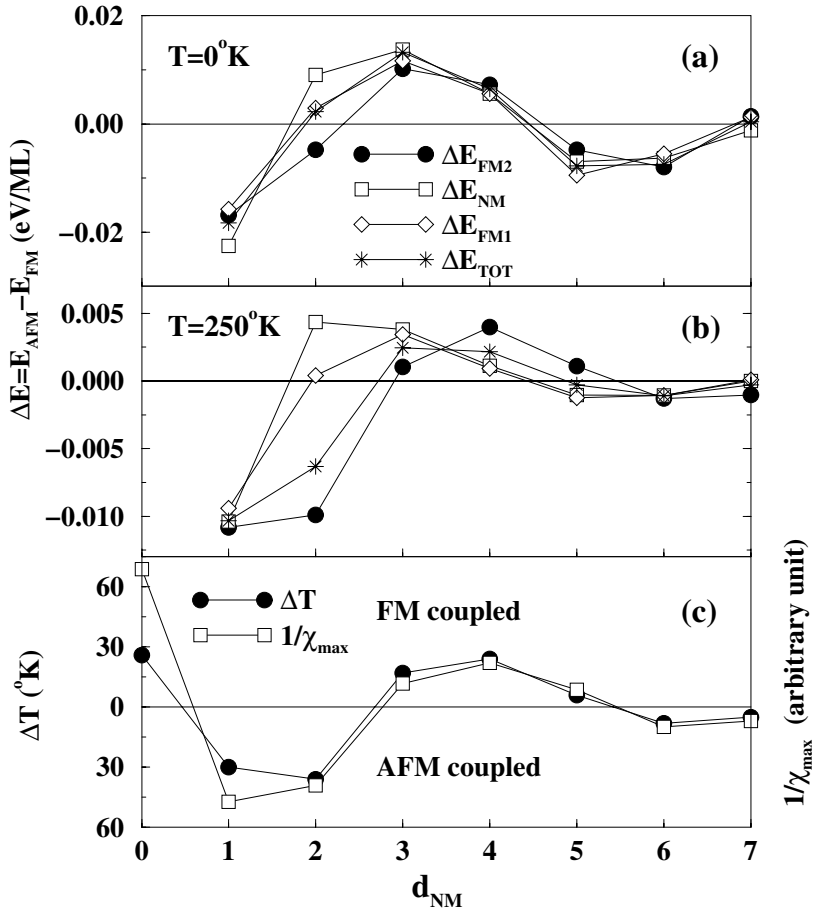

Figure 4. The energy difference ΔE at $T = 0$ K (a), $T = 250$ K (b), and the Curie temperature shift ΔT_{C2} as well as χ_{max} (c) as functions of d_{NM} in S1: $2/d_{NM}/4/6$.

Now let us turn to the relation between ΔT_{C2} and the strength of the IEC. We calculated the internal energy difference $\Delta E = E_{AFM} - E_{FM}$ between the ferromagnetic configuration (FMC: where the directions of the magnetizations of FM1 and FM2 are parallel) and anti-ferromagnetic configuration (AFMC: where the directions of the magnetizations of FM1 and FM2 are antiparallel) (figures 4(a), 4(b)). Here we would like to point out that the total-energy difference ΔE_{TOT} must include the contributions of all sublayers—FM1, FM2, and NM—not only the NM as in previous theories [14]. The energy differences of FM1 and FM2 (ΔE_{FM1} and

† Note that T^*_{C2} is different from that obtained from magnetization as a function of temperature in experiments [9, 10].

ΔE_{FM2}) are of the same order of magnitude as that of the NM (ΔE_{NM}). Furthermore, they can have different signs of ΔE_{NM}, which can even induce a coupling for the whole system which is opposite to that resulting from the NM only. In a full consideration, the energy difference of SUB (ΔE_{SUB}) should also be included, but we find that ΔE_{SUB} is very small compared with the other differences, so it can be neglected. From figures 4(a) and 4(b), we find that ΔE_{TOT} and also ΔE_{FM1}, ΔE_{FM2}, and ΔE_{NM} have oscillatory behaviours with respect to the thickness of the NM. Further investigations show that the period of this oscillation is almost the same as that of the induced polarization oscillation in the NM for the uncoupled systems [29]. This period is determined mainly by the properties of the NM, while it is hardly affected by the properties of the FMs. But the amplitude of the oscillation of ΔE_{TOT} is mainly determined by the properties of the FMs. On the other hand, we find the magnetizations of FMs are different in the FMC and AFMC. The largest difference of the mean magnetization of FM2 can reach 30% in our calculations, but that of FM1 is very small and less than 2%. This means that if we replace the FMs by a spin-dependent potential, the height of this potential must be different in the FMC and AFMC. However, the present theories [14] of the IEC always neglect this effect. From figures 4(a) and 4(b), it can also be seen that the coupling between FM1 and FM2 has changed signs in some cases ($d_{NM} = 2, 5, 7$) as temperature increases. This means that there exists a temperature-induced transition of the IEC. Comparing figure 4(c) with figures 4(a) and 4(b), we can find that ΔT_{C2} is related to ΔE_{FM2} but not ΔE_{TOT} at high temperature. Because at T^*_{C2} the unstable state cannot be obtained from our self-consistent calculation, we cannot obtain the energy difference very close to T^*_{C2}.

4. Summary

Summarizing, within the single-band Hubbard model we have investigated the change of magnetic properties in indirectly coupled systems (FM1/NM/FM2/SUB). The Coulomb interaction between electrons in the FMs is treated by using the SDA, which can lead to rather convincing results concerning the magnetic properties, especially at finite temperatures. The magnetic susceptibility is used to define the quasi-Curie temperature, and the Curie temperature shift. The relation between the Curie temperature shift and the strength of IEC is discussed.

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