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ORIGINAL PAPER

Finite Size Effect in Cu-doped Ni thin Films

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Abstract

The finite size effect power law in Cu-doped Ni thin film were investigated by probing the Curie temperature (Tc)-dependence on the Ni_xCu_{1-x} thin film thickness. It is found that both magnetization and Tc decreases proportional to the Cu content. Tc is directly proportional to the number of pairwise spin-spin interactions in a manifold cluster of spins according to the mean field approximation. The experiment results confirm the spin bag model built in ultrathin magnetic films previous, in which each spin interacts equally strongly with neighbors over some finite interaction length N_0. Furthermore, upon varying the composition of the Ni-Cu alloys, the range of spin-spin interactions N_0 decreases with increasing concentration of the Cu. Since there is little change of the band structure and conduct electron properties in Ni_xCu_{1-x} alloy films, this phenomenon was attributed to the decreasing of the propagation distance of conduct electrons due to the scattering by the Cu impurities. The present results indicate the ferromagnetic order in 3d metal probably delivered by the itinerant electrons.

Keywords Alloy films · Finite size effect · Spin-spin interactions

The understanding of the itinerant magnetism and corresponding spin-spin interaction in metallic magnets is controversial both theoretically and experimentally, spin-spin coupling mechanism in 3d transition metal is the key point in the explaining of the long range magnetic order. In order to interpret the spin correlation in magnetic metal, several models have been proposed. The Stoner theory of ferromagnetism is the earliest model which successful describe the metal magnetism behavior at 0 K, unfortunately, it led to a calculated Tc for iron of over 10^4 K. Ruderman-Kittel-Kasuya-Yoshida(RKKY) model describe the spin interaction between ferromagnetic layers across a nonmagnetic metal spacer very well [1, 2]. However, it is too weaken to explain the origin of the 3d metal magnetism, to date, it is still unclear about the spin-spin coupling mechanism in the formation of magnetic order. To make clear this issue, it need to get more information about the spin coupling interaction in metal magnetism.

It is well known that Tc is one of the most important properties to characterize FM phase stability, which is also tightly related to the spin-spin interaction of both metal and insulator magnetism. In two-dimensional thin film, finite size effect refers to the Tc will shift to lower temperatures when the spin-spin correlation exceeds the film thickness [3, 4]. The phenomenon had been observed in Fe [5], Co [6, 7], Ni [7–11], Gd [12, 13], and CuMn spin-glass films [14]. To explain the thickness dependence of the Tc in nanoscale thicknesses, a so-called "spin bag" model was employed by Zhang and Willis [15]. According to this model, the "itinerant exchange" is no longer confined to nearest-neighbor pairwise exchange interactions but can extend over longer distances, and the critical temperature Tc(n) of a thin film of n monolayers (ML) with respect to the bulk value Tc(∞) can be ascribed as equation below, (where $t = 1-\frac{T}{T_c}$ is the reduced temperature, the shift exponent λ reflects the appropriate universality class [3]).

$$t(n) = 1-\frac{T_c(n)}{T_c(\infty)} = 1-\frac{n-1}{2N_0} \quad (n < N_0) \tag{1}$$

$$t(n) = 1-\frac{T_c(n)}{T_c(\infty)} = \left(\frac{N_0+1}{2n}\right)^{\lambda} \quad (n > N_0) \tag{2}$$

From the theoretical model above [15], in the ultrathin film range (n < N_0), $t(n)$ is linear with n [Eq. (1)]. While for film thickness $n > N_0$, $t(n)$ turn to a power law curve with shift exponent λ = 1, here N_0 is the range of the spin-spin coupling.

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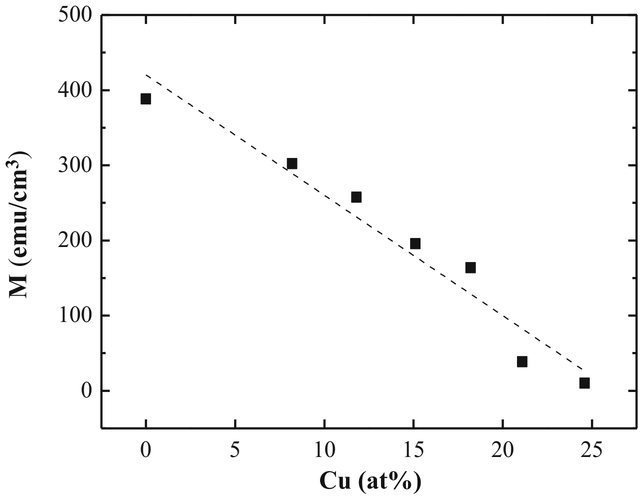


Fig. 1 The dependence of the magnetic moments for Ni_xCu_{1-x} alloys thin film on the Cu content at room temperature

This model is consistent with the data about Ni very well which interaction range N_0 can be obtained closely to the 5 ML, namely, the spin-spin coupling range of the Ni itinerant magnetism exceed the nearest-neighbor exchange interactions. Apparently the associated parameter N_0 can provide important information about the origin of the spin coupling in magnetic metal film systems.

By testing thickness dependence of Curie temperatures Tc(n) in ultrathin film, N_0 can be obtained directly by identifying the demarcation between the power law and linear behavior, since the thicker films ($n > N_0$) show a power law dependence with $\lambda = 1$, and the thinner films ($n < N_0$) show a linear dependence on thickness. Unfortunately, Tc can be influenced prominently in the presence of adjacent bottom and capping layer [16, 17]. Moreover, the ferromagnetic layer should be affected in the course of the Tc measurement, in

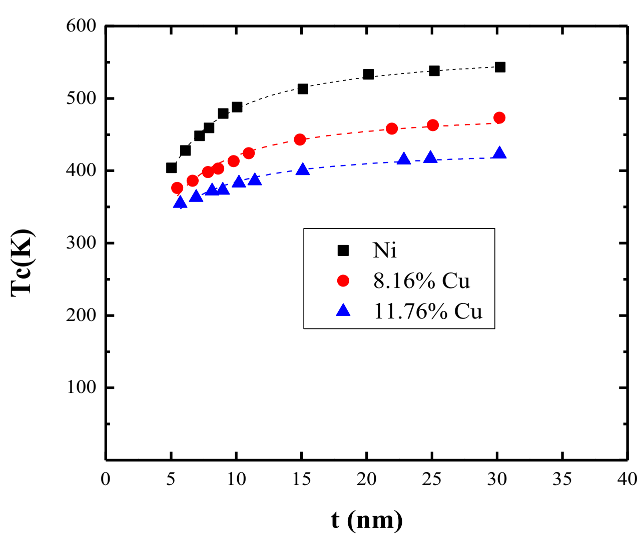


Fig. 2 The dependence of Curie temperature on the thickness for the Ni (black), $Ni_{92}Cu_8$ (red) and $Ni_{88}Cu_{12}$ (green) alloys thin film

which high temperature will lead to interface inter-diffusion, especially in ultrathin film which can be formed an alloy layer at the interface. In this case, it is essential to investigate the N_0 behavior in ferromagnetic alloy film.

Very recently, 3d transition metal doped into two-dimensional materials has attracted a great deal of attention for their rich physics and important applications in spintronics devices [18–20], it undoubtedly can provides a new insight in studying the mechanism of spin-spin coupling as well. It had been reported that the alloy composition of transition-metal film can affect N_0. However Tc is sensitive to the electronic properties as well as crystal structure, upon varying the composition of the alloys, some properties of electron may also been altered, such as the electronic band structure, the mean free path of the conducting electrons as well as the distribution of asymmetric spin-polarization electrons at the Fermi surface in magnetic metal [21]. Therefore, it is difficult to obtain the intrinsic properties of the spin-spin interaction range without considering these factors.

Fortunately, the Ni_xCu_{1-x} alloy is an ideal ferromagnetic alloy to study parameter N_0 by using finite size effect. Firstly, it has already been verified that as the Cu-doped in the Ni, the 4 s electrons of Cu atoms enter the Ni 3d orbits without modifying the Ni outer s electron, and the electron properties at Fermi surface change little with the variation of the Cu composition in Ni_xCu_{1-x} [22]; secondly, Ni and Ni_xCu_{1-x} alloy are the “weak ferromagnets” which have rather low bulk Tc, and N_0 can be determined from Eq. (2) by probing the Tc-dependence on the thickness range of $n > N_0$. In this paper, we study the intrinsic nature of spin-spin interaction range N_0 by using finite-size scaling behavior in Ni_xCu_{1-x} alloy films. To minimize the disturbance of the inter-diffusion from the neighboring layer at the interfaces in high temperature [23], all samples thickness were prepared from 5 nm to 30 nm.

Samples were deposited in magnetron sputtering system with base pressure of 5×10^{-5} Pa. To induce a uniaxial anisotropy in the FM layer, a magnetic field of about 200 Oe was applied parallel to film plane along the long axis of the sample during growth. Magnetic properties were measured by vibrating sample magnetometer (VSM).

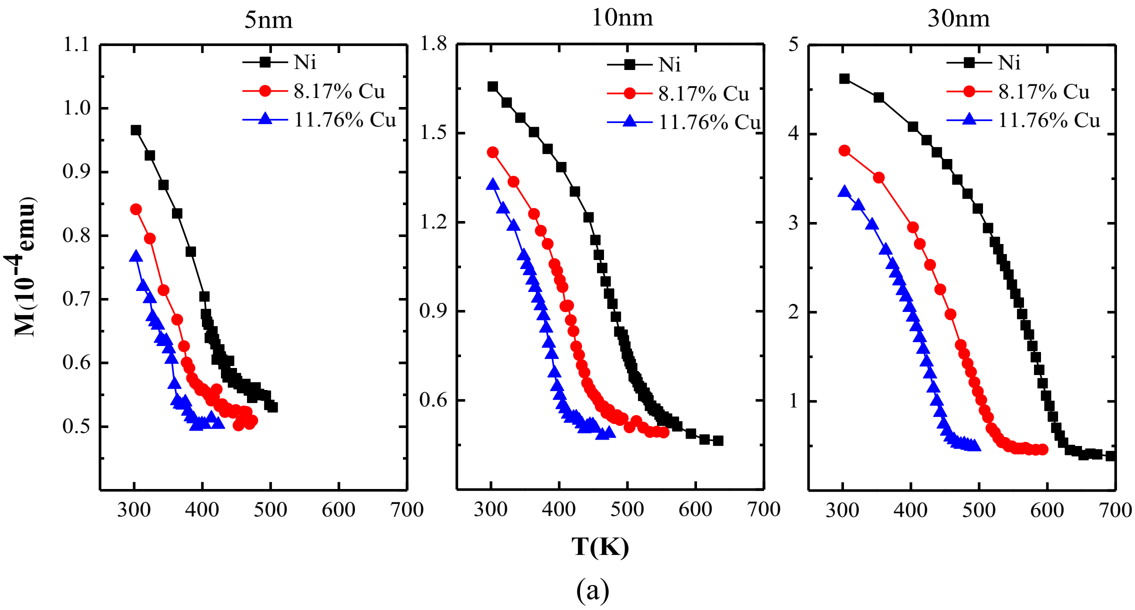
Samples with stack of Cu(5 nm)/Ni(t)/Cu(5 nm) and Cu(5 nm)/Ni_xCu_{1-x}(t)/Cu(5 nm) have been grown on the glass substrate by a multisource sputter deposition system, where the bottom and top Cu layer is used as buffer layer and capping layer, respectively. The Ni_xCu_{1-x} alloy was made by co-deposition from two separate sources of Ni and Cu, the composition of Ni_xCu_{1-x} alloy was tuned by adjusting the deposition rates of the Cu, and each sample was cut into the same size exactly by dicing machine.

Figure 1 shows the dependence of the magnetic moments of 20 nm Ni_xCu_{1-x} film alloys on the Cu content, one can find that the magnetization of the Ni_xCu_{1-x} film decreasing approximately linear with increasing of the Cu compositions. As mentioned early, since the Cu impurities only affect the local Ni atoms inner

3d orbit without altering the electronic properties of the conductor electrons, as a result, the magnetization of the Ni atoms should decrease proportional to the Cu contents, where is agreement with our experimental results very well.

Two series Ni_xCu_{1-x} alloys thin film samples with thickness range from 5 nm to 30 nm were prepared, where the compositions of the Cu are 8.16% and 11.76%, respectively, as the contents of the Cu is further increasing, the magnetic moments signal of the Ni_xCu_{1-x} thin film is too weak to detectable by the VSM. Figure 2 shows the dependence of Curie temperatures on the Ni_xCu_{1-x} alloy layer thickness for different compositions of Ni_xCu_{1-x}. Tc shows not only a decline with decreasing thickness, but also decrease with the increasing of the Cu content.

According to the model based on the mean field approximation by Renjun Zhang and Roy F. Willis,



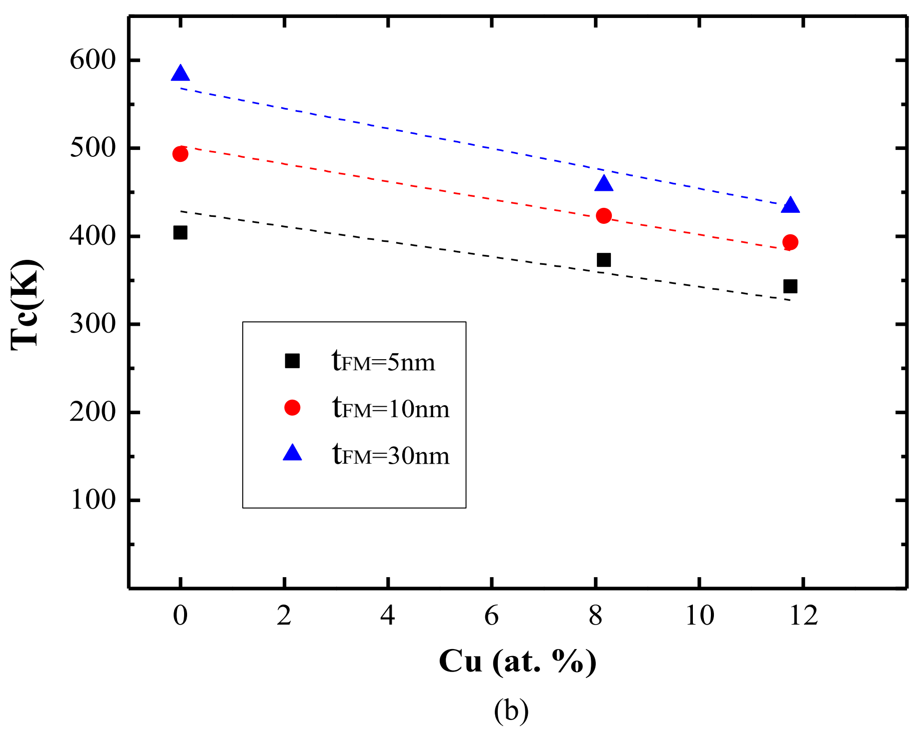


Fig. 3 **a**, Magnetization(M)-T curve of Ni_xCu_{1-x} alloy film with 5 nm,10 nm and 30 nm, respectively. **b** The dependence of Curie temperature on the 5 nm, 10 nm, and 30 nm thickness for the Ni_xCu_{1-x} alloys with varying Cu content, the black, red, and blue line are the guide to the eye

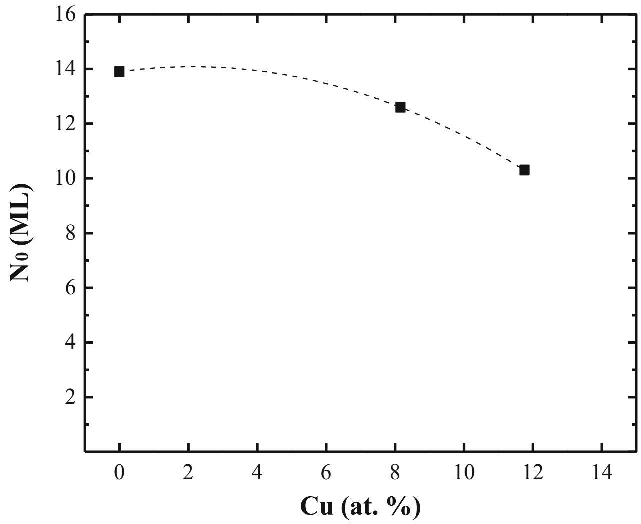


Fig. 4 The dependence of the spin-spin interaction range $N0$ for Ni_xCu_{1-x} alloys thin film with varying Cu content. The fitting parameter $N0$ is deduced using Eq. (2)

$$k_BT_c = N \times E_0 \tag{3}$$

where N is the number of pairwise interactions and E_0 is a constant ("the coupling energy"), the Tc is proportional to the spin number within the coupling range N_0. When Cu is doped into the Ni film, the outside 4s electrons of Cu atoms will enter into the 3d unfilled orbit of Ni, and the full 3d atoms orbit will make it lose local moment. At the same time, the Ni outer s electron states and band structure change little with the Cu composition in Ni_xCu_{1-x} alloy, therefore, the coupling energy E_0 in each pairwise can be assumed unchanged upon Cu doping. In this case, each Cu-substituted atom inside the Ni_xCu_{1-x} alloys will damage two pairwises spin interactions. According to the Eq. (3), 8.16% and 11.76% Cu content in Ni_xCu_{1-x} alloys of present work should lead to the Tc value decrease approximate 16% and 23%. Figure 3(a) show the magnetization(M)-T curve of Ni_xCu_{1-x} alloy film with 5 nm,10 nm, and 30 nm thickness, respectively, here Tc were arrived at where dM/dT exhibits a minimum [24]. Figure 3(b) show the dependence of the Tc on the Cu content, obviously the agreement of the experimental date with the eq. (3) is very well.

The relationship between the itinerant electrons and local spin moments during the formation of the ferromagnetic order is important to understand 3d metal magnetism. The behavior of spin-spin coupling range N_0 in Ni_xCu_{1-x} can be regarded as the key of this issue. With this in mind, the dependence of N_0 on the Cu content in Ni_xCu_{1-x} alloys was calculated from Eq. (2). Ni lattice constant of 0.2 nm was adopted to fit the N_0-Cu% curve, as shown in Fig. 4. It was found that N_0 decreases as the Cu compositions increasing, indicates the spin-spin interaction range N_0 decrease progressively with the Cu content.

As mentioned above, the outside 4s electrons of Cu atoms in Ni_xCu_{1-x} give rise to a decreasing of localized moments and magnetization of the ferromagnetic film merely, as shown in Fig. 1, in which the spin magnetic coupling between local electrons are unaffected by the Cu impurities because the band structure remain unchanged. We point out that if the effective range of the spin-spin interactions result from the local d or sp-d hybridization, the parameter N_0 are invariant. Thus, a conclusion can be drawn that the decreasing of N_0 present here probably not origin from the local electron. We assume that the electrons which delivering the spin-spin interaction in a metal are rather free to travel through the thin film, and N_0 imply the spin coupling effective range that the electrons can reach to, which is decided not only by the energy band structure but also affected by the impurity scattering. In this case, each Cu atom can be regarded as a impurity scattering source which shorten the propagation distance of the deliver electrons, finally led to the decreasing behavior of N_0 in Ni_xCu_{1-x} alloy film. The work support a itinerant electrons deliver ferromagnetism coupling mechanism in Ni metal.

In summary, we have investigated the dependence of Tc on the thickness of Ni and Ni_xCu_{1-x} alloy films. By studying the Ni_xCu_{1-x} alloy Tc-dependence on the Cu content in detail, it is found that Tc is proportional to the number of the spin pairwise interactions within the spin-spin coupling range, which agreement with the mean field approximation very well. More important, the spin-spin interaction range N_0 was observed decay with the increasing of the Cu contents in Ni_xCu_{1-x} alloys. We proposed a explanation that the attenuation of the N_0 is due to the reduction of electrons "magnetic mean free path" by the Cu impurity scattering. The work provide a physics picture that the formation of long-range magnetic order of metal magnetism probably originate from a non-Heisenberg coupling interaction, in which the spin-spin coupling are delivered by the conducting electrons. This work possibly shed light on the mechanism of the formation of the 3d metal ferromagnetic phase, and the itinerant magnetism should be built on a more freely electrons interaction model.

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