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# Random interaction fields method: Magnetic phase transitions in the thin films

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ABSTRACT

In the framework of the random interaction fields method the properties of the magnetic thin films with non-magnetic layers are investigated. In this paper the evaluation of the interaction energy of Co layers and the oscillation period is carried out, depending on the thickness of the nonmagnetic layer Cu, assessing of the impact of the cobalt layer thickness on the average effective field RKKY interaction. Further, a possible explanation of the superparamagnetic properties of such systems is offered.



## 1. Introduction

In recent years, the physical phenomena in a periodic structures consisting of the alternating layers of different metals have been widely studied theoretically [1–3] and experimentally [4,5]. The interest in such systems is large because the thin film elements used in microelectronics are often representing multilayer system [6,7]. On the other hand, the metal films having different physical properties can be combined, then it is possible to obtain wires with fundamentally new physical properties which cannot be implemented in a homogeneous conductor. Thus, in particular, multilayers consisting of the alternating layers of magnetic and non-magnetic metals have giant magnetoresistance, which manifest themselves in changing the conductivity by tens of percent when there is the weak external magnetic field [8–10]. This effect is perhaps the most striking example of the unusual properties of nanostructured materials. The systems of Co/Cu and Co/Pd are most interesting in this field. Extremely low mutual solubility of the components of these systems allows us to obtain well-differentiated layers or other type of structure in the film version. The unusual magnetic properties, such as spin glass state [6] or quasicrystals [7], and size effects in ultrathin films [11,12] can be observed. It explains the increased interest and a huge amount of work on the films of Co/Cu and Co/Pd of different compositions which exist in the scientific journals. However, some experimental works have difficulties in interpreting results within existing theories, different experiments demonstrate various oscillation periods, etc.

Apparently, the direct exchange determines the Curie point and the temperature behavior of the spontaneous magnetization of Co, while relatively weaker RKKY interaction between spins of cobalt layers affects the relative orientation of the magnetization vectors and determines the energy of the interlayer interaction. In our view, many of the properties of such structures can be consistently explained using the random interaction fields method developed in our studies [13–18].

## 2. Evaluation of the interaction energy of cobalt layers and the oscillation period, depending on the thickness of the nonmagnetic layer

As was shown in [13–18], the distribution function of random fields $W(H)$ is "smeared" $\delta$-function of the form

$$W(H)=\frac{1}{\sqrt{\pi}B}\exp\left(-\frac{[H-H_0(\alpha-\beta)]^2}{B^2}\right), \tag{1}$$

where the moments of the distribution function can be represented as

$$H_0=n\int \varphi\, dV, \tag{2}$$

$$B^2=2\sigma^2=2n\int \varphi^2\, dV. \tag{3}$$

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Here $(\alpha-\beta)$ is the relative magnetic moment per atom, $\varphi$ is the "exchange field strength" acting on the selected atom from the neighboring atom, $n$ is the volume concentration. "Exchange field strength" of alternating indirect Ruderman–Kittel–Kasuya–Yosida (RKKY) law is as follows:

$$\varphi = AF(x), \tag{4}$$

where $x = 2k_F R$ is the momentum of the electron on the Fermi surface

$$F(x) = \frac{x\ \cos(x) - \sin(x)}{x^4}, \tag{5}$$

the magnitude $A \approx 10^6$ Oe determines the intensity of the exchange interaction (estimation of the magnitude $A$ is based on the evaluation of the Curie point $T_c \approx 200$ K).

Due to the long-range nature of the RKKY interaction, when a substantial contribution to the interaction comes from the atoms located at the distances larger than the lattice constant, the distribution of the interacting atoms in the volume can be considered as random. Then its moment can be determined as $H_0 = n\int \varphi(r)\,dV$, here $r$ is the coordinate of the atom acting on the selected atom placed at the origin.

If the coordinate system is associated with the surface layer of Co thickness $h$, then the part of the average exchange field per atom Co, located at the origin, associated with the RKKY interaction should be calculated using the following formula:

$$H_0 = nA\int_{-h}^{0}\int_0^{\infty}\int_0^{2\pi} F(\rho,\varphi,z)\rho\,d\rho\,d\varphi\,dz + nA\int_d^{d+h}\int_0^{\infty}\int_0^{2\pi} F(\rho,\varphi,z)\rho\,d\rho\,d\varphi\,dz, \tag{6}$$

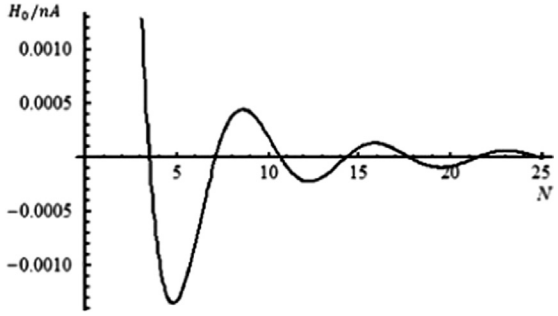


**Fig. 1.** Dependence of $H_0/nA(N)$ where $N$ is the monolayers of Cu (1 monolayer=2 Å) at 1 monolayer of Co.

where $\rho, \varphi, z$ are the cylindrical coordinates. It is possible to obtain simple results by assuming that the law of the RKKY interaction between the atoms through the Co–Co layer is either absent or is not very different from the law of interaction through the layer of copper. Then, considering only the interaction of the surface the atoms of Cu with the atoms Co Eq. (6) is transformed into the form

$$H_0 = nA\int_0^{\infty}\int_0^{2\pi} F\sqrt{\rho^2+d^2}\,\rho\,d\rho\,dz, \tag{7}$$

where $n$ is number of atoms per unit surface layer Cu (Pd), $d$ is the thickness of the layer Cu (Pd). The dependence of the magnitude $H_0/nA$ on the thickness of Cu is shown in Fig. 1 (1 monolayer of Cu=2 Å ) with a layer thickness of Co equal to 1 monolayer.

Note that the experimental curve can look quite differently at a certain step in increasing the thickness of the copper layer. For example, choosing $N$=2; 4; 4.5; 6; 6.5; 7; 8; 10, we obtain the dashed curve shown in Fig. 2a compared with experimental data obtained in [18], similar to Fig. 2b.

We can get to the points lying in the region of negative values of the exchange integral and can have oscillating antiferromagnetic ordering at such a discrete change of the intervals (Fig. 2a).

## 3. Estimation of influence the cobalt layer thickness on the average effective field RKKY interaction

In order to evaluate the influence of the cobalt layer thickness on the average effective field RKKY interaction, it is assumed that the main characteristics of the RKKY interaction in Co and Cu are comparable in the order of the magnitude. In this case, we use Eq. (6), which when integrated from $d$ to $d+h$ functions $F(\rho, d)$ are identical in the region occupied by the copper and the cobalt.

Effect of Co layer should not be considered in the range of $-h$ to 0 when evaluating $H_0$, because the magnitude $H_0$ together with the direct exchange specifies a Curie point in a layer Co:

$$H_0 = A\int_d^{d+h}\int_0^{\infty}\int_0^{2\pi} F(\sqrt{\rho^2+z^2})\rho\,d\rho\,d\varphi\,dz. \tag{8}$$

The corresponding graph is shown in Fig. 3 in which the thicknesses of Co are 20 monolayers (smooth line), 10 monolayers (dotted line), and 5 monolayers (dashed line).

When comparing Figs. 1 and 3, it is seen that the order of the magnitude of the mean field of the exchange interaction between the layers and the oscillation period is not changed.

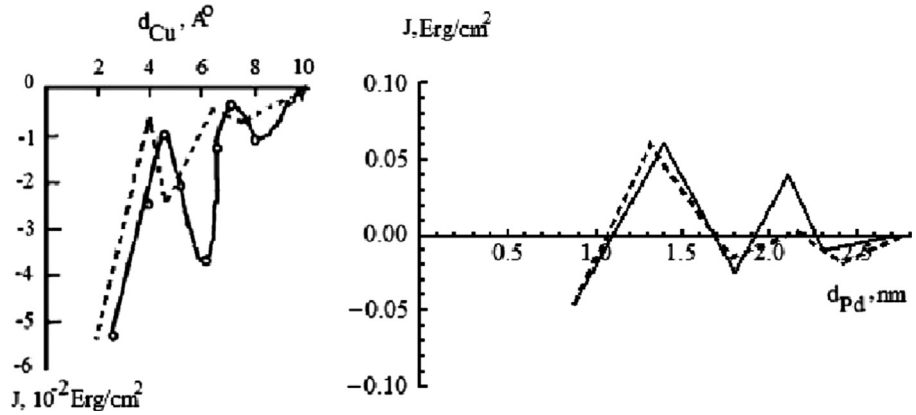


**Fig. 2.** (a) Plots of the dependence interlayer exchange interaction on the thickness Cu in the films CoCu. Black curve was obtained by means of the experimental data [18], dashed curve was obtained by means of the theoretical data in the framework of the random interaction fields method. (b) Plots of the dependence interlayer exchange interaction on the thickness Pd in the films CoPd. Black curve was obtained by means of the experimental data [19], dashed curve was obtained by means of the theoretical data in the framework of the random interaction fields method.

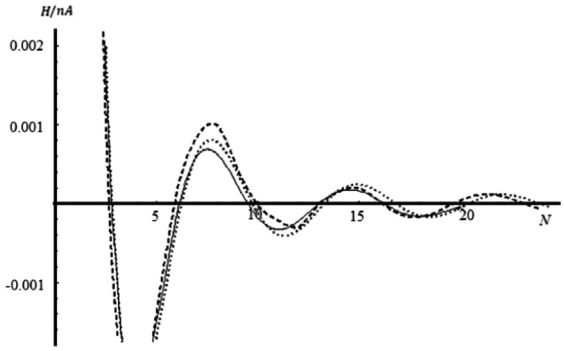


**Fig. 3.** Dependence of $H(N)$, where $N$ are monolayers of Cu (1 monolayer = 2 Å) at a thickness of 20 monolayers of Co (smooth line), 10 monolayers Co (dotted line), 5 monolayer Co (dashed line).

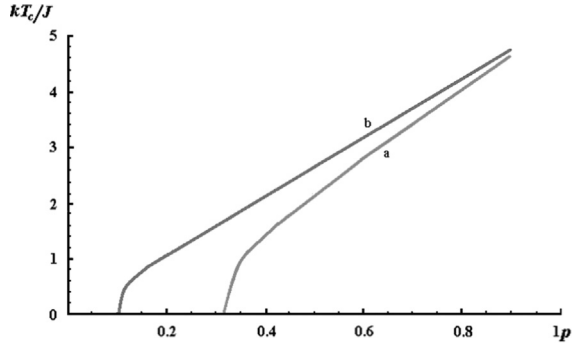


**Fig. 4.** Dependence of $kT_c/J(p)$. Curve (4a) illustrates the absence of the diffusion, curve (4b) illustrates that the diffusion is present.

## 4. Superparamagnetism films

Based on an analysis of the certain experimental data one can conclude that the cobalt layer is a band of superparamagnetic particles [18]. As an alternative explanation of the behavior of a material in a magnetic field can make the assumption that there has been partial mixing of Co and Cu atoms the diffusion leads to areas with high concentrations of Co, acting as superparamagnetic particles. In this case of an arbitrary node of the crystalline lattice of atoms at a concentration of Co $p < 1$ there are three possible states:

1. Site is occupied by a ferromagnetic atom with spin oriented "up".
2. Site is occupied by a ferromagnetic atom with spin oriented "down".
3. Site is occupied by non-ferromagnetic atoms.

Then the grand partition function is

$$Z = 1 + \lambda \exp\left(\frac{m_0 H_0}{kT}\right) + \lambda \exp\left(-\frac{m_0 H_0}{kT}\right), \tag{9}$$

where $\lambda = \exp(\mu/T)$, $\mu$ is the chemical potential, $m_0$ is the magnetic moment per atom. The average value of the magnetic moment $pM$ is determined from the formula:

$$\langle pM \rangle = \int \frac{2\lambda \sinh\left(\frac{m_0 H_0}{kT}\right)}{1 + 2\lambda \cosh\left(\frac{m_0 H_0}{kT}\right)} W(H_0)\, dH_0, \tag{10}$$

the concentration of non-ferromagnetic atoms is

$$1 - p = \int \frac{W(H_0)}{1 + 2\lambda \cosh\left(\frac{m_0 H_0}{kT}\right)} dH_0. \tag{11}$$

In the following equations $\beta = 1/2\lambda, m_0 = 1, k = 1$ the exchange integral is also equal to one.

Thus, the Curie temperature as a function of the concentration of $p$ can be found from the simultaneous solution of Eqs. (10) and (11).

As follows from Eq. (10)

$$\langle pM \rangle^2 = \frac{6\left(\frac{H_0 \sinh\left(\frac{B}{T}\right)}{B\left(\beta + \cosh\left(\frac{B}{T}\right)\right)} - 1\right)}{\frac{H_0^3\left(2 - \beta^2 + \beta \cosh\left(\frac{B}{T}\right)\right)\sinh\left(\frac{B}{T}\right)}{BT^2\left(\beta + \cosh\left(\frac{B}{T}\right)\right)^3}}. \tag{12}$$

The present expansion of the small parameter is $pM$, but is not $M$, as in Eq. (8), therefore $H_0$ does not include the factor $p$. As follows from Eq. (11),

$$\frac{\frac{\sqrt{\beta^2 - 1}\tanh\left(\frac{B}{2T}\right)}{1 + \beta}}{\tanh\left(\frac{(1-p)B\sqrt{\beta^2 - 1}}{2\beta T}\right)} = 1. \tag{13}$$

Curie point – concentration of "ferromagnetic" atoms diagram for a cubic lattice ($z = 6$) – is shown in Fig. 4. Curve (4a) illustrates the absence of the diffusion, curve (4b) illustrates that the diffusion is present.

The presence (curve 4a) of the critical concentration of $p_c = 2/z$ indicates that there is not the "flowing cluster" for frozen impurities at $p < p_c$, and there is the spin glass state at the decreasing temperature. If the magnetic impurity can come in thermodynamic equilibrium with the system, the diffusion can influence to the formation of the "flowing cluster" at $p < p_c$. It was noted in the studies of ultrathin films earlier [20–22]. As this process evolves in time, the Curie temperature can grow in the presence of an external magnetic field, it can cause a stable remanent magnetization, the mechanism of which is connected with the passage from the Curie point $T_c < T$ to $T_c > T$. A similar transition is known which leads to the formation of the so-called thermoremanent magnetization, with the only difference that in this case when the magnitude of $T_c$ is static the temperature drops from $T > T_c$ to $T < T_c$. It is also obvious that the diffusion leads to the formation of the areas with high concentration of magnetic atoms on the background areas of their low content.

## 5. Conclusion

Thus, the properties of the magnetic films with the non-magnetic layers are explained in the framework of the random interaction fields method:

1. If we take the experimental points on the theoretical diagram and link them we can obtain a continuous curve, and this graph has the same form as that of the experimental graph. It indicates that the experimentally determined oscillation periods cannot correspond to reality.
2. Assessing the impact of the thickness of the cobalt layer on the average effective field RKKY interaction shows that the order of the magnitude of the mean-field exchange interaction between layers and the oscillation period is not changed.
3. Moreover, using a two-sublattice model of a magnet and allowing the possibility of diffusion of atoms between Co and

Cu found areas with the high concentrations of Co, acting as superparamagnetic particles.

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