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Magnetic Studies of Spin Wave Excitations in Ni/Cu Multilayers

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Abstract

The magnetic properties of Ni/Cu multilayers, prepared by the electron beam evaporation method under ultra high vacuum conditions, have been systematically studied by magnetic measurements. The temperature dependence of the spontaneous magnetization M (T) is well described by a $T^{3/2}$ law. A spin wave theory has been used to explain the magnetization versus temperature. Based on this theory, the approximate values for the exchange interactions have been obtained.

Keywords: Ni/Cu Multilayer, Magnetization, Spin Wave Excitations, Exchange Interactions

1. Introduction

Magnetic multilayers with artificial periodicity constitute a topic of active investigations, both on the fundamental research level as well as on the applied one. Several experimental and theoretical studies were carried out to understand the origin of the various interactions existing in this kind of systems, in particular in the interface since it presents magnetic properties different those of bulk [1-4]. Since multilayers are inherently metastable material on a nanometer scale, the introduction of the period, the number of layers and the relative thicknesses of the magnetic layers and nonmagnetic layers in multilayers will result in many interesting properties, which are sensitive to the microstructures [5-8]. The properties of these materials are mostly governed by the surface properties and hence the interface plays an important role. The discovery of coupled magnetic behavior between layer components in various magnetic multilayer systems has led to an increased interest in two-dimensional systems. To understand how the interlayer magnetic coupling between ferromagnetic layers through nonmagnetic layers affects the magnetic dynamics of such a coupled magnetic system, we carried out an investigation on a multilayer system. Furthermore, due to the immiscibility of Cu and Ni, the Ni/Cu system is an excellent one to investigate with nearly ideal artificially structures Ni/Cu multilayers with flat and sharp layer interfaces are possible using the electron beam evaporation.

In this paper we study the properties of spin waves in Ni/Cu multilayers prepared by evaporation under ultra high vacuum conditions. The comparison between the calculated and experimental magnetization enabled us to make a satisfactory estimate of the various exchange integral values.

2. Experimental

Ni/Cu multilayer was grown by evaporation in ultrahigh vacuum under controlled conditions using several electron guns. The pressure during the film deposition was maintained in the range 3 - 5 × 10^{-9} Torr. The deposition rate (about 0.3 Å/s) and the final thickness were monitored by precalibrated quartz oscillators. All the samples were deposited on glass substrate at 300 K on a nonmagnetic buffer layer 100 Å thick. The Ni-layer thickness t_{Ni} was varied from 14 to 50 Å and that of t_{Cu} were kept fixed at 20 Å.The number q of bi-layers were in the range 10 - 30. The x-ray diffraction in the high angle range 35˚ < 2θ < 50˚ showed the existence of fcc Ni (111) peak. Magnetization M was measured using a vibrating sample magnetometer under magnetic fields up to 1T and in the temperature range 5 to 650 K.

3. Results and Discussion

The magnetization depended strongly on both the Ni and Cu layer thickness, for such thicknesses of the order of a few atomic planes. The low-temperature magnetization (per unit volume of the Ni content) was found to reach values up to about 15% higher than that of pure Ni, due to an enhancement of the Ni moment in Ni/Cu multilayer. **Figure 1** shows the temperature dependence of M for several values of t_{Ni} thicknesses. It can be noticed that the Curie temperature T_C decreases with decreasing Ni thickness due to reduced coordination. For three-dimensional magnetic films, the magnetization has a $T^{3/2}$ dependence (for temperatures as high as $T_C/3$) due to the classical spin-wave excitations. In such cases, according to spin-wave theory, the temperature dependence should follow the relation:

$$M(T) = M(5K)\left(1 - BT^{3/2}\right) \quad (1)$$

This behavior is observed for temperatures less than $T_C/3$.

The spin-wave constant B decreases from 86 × 10^{-6} $K^{-3/2}$ for t_{Ni} = 14 Å to 30 × 10^{-6} $K^{-3/2}$ for t_{Ni} = 50 Å for Ni/Cu multilayer. It is seen that B is much larger than the value of 7.5 × 10^{-6} $K^{-3/2}$ found for bulk Ni.

The B versus $1/t_{Ni}$ is plotted for the samples with 14 ≤ t_{Ni} ≤ 50 Å in **Figure 2**. It is seen that the experimental points align well in a straight line. The values extrapolated to $1/t_{Ni} = 0$ are in good agreement with those found for the bulk Ni. It was observed that the parameters B in Equation (1) depend on t_{Ni} according to:

$$B = B_\infty + \frac{B_S}{t_{Ni}} \quad (2)$$

Where B_∞ is the bulk spin-wave parameter of Ni and B_S is the surface spin-wave parameter value.

As the saturation magnetization decreases with decreasing layer thickness, so does the Curie temperature T_C. These values of T_C are much smaller than the bulk Ni value of 630 *K*. For ferromagnetic films the decrease of $T_C(t_{Ni})$ as the layer thickness t_{Ni} is reduced and is usually described in the framework of finite-size scaling, which predicts in Equation (3) that $T_C(t_{Ni})$ scales with t_{Ni} via a shift exponent λ [9]

$$\frac{T_C(\infty) - T_C(t_{Ni})}{T_C(\infty)} = C \cdot t_{Ni}^{\lambda} \quad (3)$$

Where *C* is a constant depending on the nature of the material.

The fit further results (**Figure 3**) in a "shift exponent"

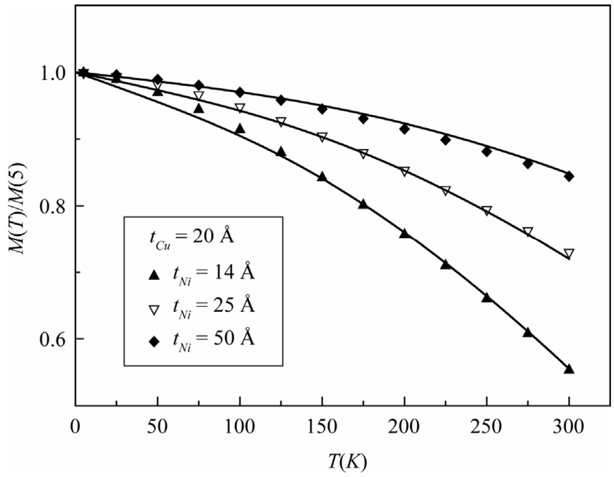

Figure 1. Calculated (continuous line) and measured (symbols) temperature dependence of the normalized magnetization of Ni/Cu multilayers versus Ni thicknesses.

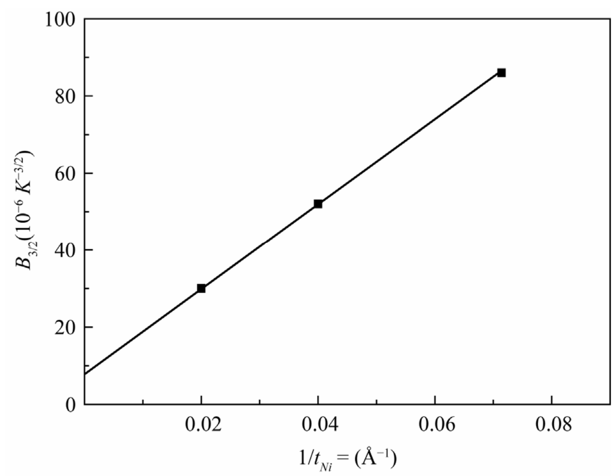

Figure 2. The spin wave constant B dependence of Ni/Cu multilayers versus inverse of Ni thicknesses.

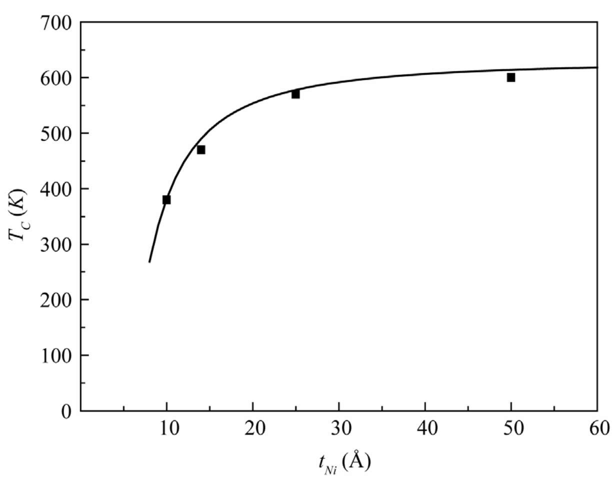

Figure 3. Curie temperature as a function of film thickness t_{Ni}.

λ = 1.7, which should be connected with the critical exponent v of the correlation length by $\lambda = 1/v$ [9,10]; our results, obtained from multilayers, are in accordance with the value $v = 0.72$ of the 3D Ising model.

To understand better how the exchange coupling between neighboring Ni layer affects the magnetic behavior of these films, we extended the model for spin waves in ferromagnetic thin films proposed by Pinnettes and Lacroix [11] to the ferromagnetic/nonmagnetic multilayers case. Here the multilayer $(X_n/Y_m)_q$ is supposed to be formed by an alternate deposition of a magnetic layer (X) and nonmagnetic one (Y). The multilayer is characterized by the number q of bi-layers (X/Y), the number n of atomic planes in the magnetic layer μ and the number m of atomic planes in the nonmagnetic layer. We chose the lattice unit vectors ($\boldsymbol{e}_X$, $\boldsymbol{e}_Y$, $\boldsymbol{e}_Z$) so that $\boldsymbol{e}_Z$ is perpendicular to the atomic planes. We note by $\boldsymbol{S}_{i\alpha\mu}$ the spin operator of the atom $i\left(i=1,2,\cdots,N\right)$ in the plane $\alpha\left(\alpha=1,2,\cdots,n\right)$ of the magnetic layer $\mu\left(\mu=1,2,\cdots,q\right)$.

The system Hamiltonian is given by: $H = H_e + H_a$,

He describes the exchange interactions in the same magnetic layer (bulk and surface) as well as the exchange interactions between adjacent magnetic layers:

$$H_e = -J_b\left[\sum_{\langle i\alpha\mu,j\alpha\mu\rangle}^{b}\boldsymbol{S}_{i\alpha\mu}\boldsymbol{S}_{j\alpha\mu}+\sum_{\langle i\alpha\mu,j\alpha'\mu\rangle}\boldsymbol{S}_{i\alpha\mu}\boldsymbol{S}_{j\alpha'\mu}\right] - J_s\sum_{\langle i\alpha\mu,j\alpha\mu\rangle}^{S}\boldsymbol{S}_{i\alpha\mu}\boldsymbol{S}_{j\alpha\mu} - J_I\sum_{\langle i\alpha\mu,j\alpha''\mu''\rangle}^{I}\boldsymbol{S}_{i\alpha\mu}\boldsymbol{S}_{j\alpha''\mu''} \quad (4)$$

where J_b and J_S are the bulk and surface exchange interactions. J_I is the interlayer coupling strength which depends on the number m of atomic planes in the nonmagnetic layer. The contribution of the surface anisotropy is given by:

$$H_a = D^{\perp}\sum_{i\alpha\mu}^{s}\left(S_{i\alpha}^{Z}\right)^2 + D''\sum_{i\alpha\mu}^{s}\left(\left(S_{i\alpha}^{X}\right)^2-\left(S_{i\alpha}^{Y}\right)^2\right) \quad (5)$$

where $D^{\perp}$ and $D^{//}$ are the surface anisotropy parameters for the uniaxial out of plane and in plane components, respectively, and $D_{eff}^2 = D^{\perp 2} + D^{//2}$. $D_{eff}\left(K\right) = \frac{K_S a^2}{k_b}$, where a is the lattice constant and k_b is the Boltzmann constant.

Further we denote by Σ^{Ξ} the summation on the sites of the bulk layer planes ($\Xi = b$), surface layer planes ($\Xi = s$) or the surfaces planes coupled via the nonmagnetic layer ($\Xi = I$). The symbol < > denotes the pairs of nearest-neighbors atoms or adjacent magnetic planes.

In the Holstein-Primakoff formulation [12], the creation and annihilation operators ($a_{i\alpha\mu}$ and $a_{i\alpha\mu}^{+}$) for each atomic spin are related to the spin operators by:

$$\begin{cases} S_{i\alpha\mu}^{Z} + iS_{i\alpha\mu}^{X} = \left(2S\right)^{\frac{1}{2}} f_{i\alpha\mu}\left(2S\right) a_{i\alpha\mu} \\ S_{i\alpha\mu}^{Z} - iS_{i\alpha\mu}^{X} = \left(2S\right)^{\frac{1}{2}} a_{i\alpha\mu}^{+} f_{i\alpha\mu}\left(2S\right) \end{cases} \quad (6)$$

In the frame work of non interacting spin wave theory, the linear approximation of the Holstein-Primakoff method is sufficient to describe the main magnetic behavior and the correction terms are quite-small at low temperatures ($T < T_C/3$) [3,7]. So, the value of $f_{i\alpha\mu}\left(2S\right)$ is fixed to 1.

We replace the atomic variables ($a_{i\alpha\mu}$, $a_{i\alpha\mu}^{+}$) by the magnon variables ($b_{k\alpha\mu}$, $b_{k\alpha\mu}^{+}$) after a two-dimensional Fourier transformation. It gives:

$$\begin{aligned} H = {} & H_0 + A\sum_{\boldsymbol{k}\alpha\mu}^{S}\left(b_{\boldsymbol{k}\alpha\mu}b_{-\boldsymbol{k}\alpha\mu} + b_{\boldsymbol{k}\alpha\mu}^{+}b_{-\boldsymbol{k}\alpha\mu}^{+}\right) \\ & + \sum_{\boldsymbol{k}\alpha\mu}^{S} B_k b_{\boldsymbol{k}\alpha\mu}^{+}b_{\boldsymbol{k}\alpha\mu} + \sum_{\boldsymbol{k}\alpha\mu}^{b} C_k b_{\boldsymbol{k}\alpha\mu}^{+}b_{\boldsymbol{k}\alpha\mu} \\ & + \sum_{\boldsymbol{k}(\alpha\mu,\alpha'\mu)} D_k b_{\boldsymbol{k}\alpha\mu}^{+}b_{\boldsymbol{k}\alpha'\mu} + \sum_{\boldsymbol{k}(\alpha\mu,\alpha''\mu'')}^{I} E_k b_{\boldsymbol{k}\alpha\mu}^{+}b_{\boldsymbol{k}\alpha''\mu''} \end{aligned} \quad (7)$$

where:

$$A = \frac{S}{2}\left(D^{\perp} - D^{//}\right)$$

$$B_k = 2S\left(J_s\left(n'' - \lambda_k\right) + J_b n_S^{\perp} + J_I n^{\updownarrow}\right) + S\left(3D^{//} + D^{\perp}\right)$$

$$C_k = 2J_b S\left(\left(n'' - \lambda_k\right) + n_V^{\perp}\right)$$

$$D_k = -J_b S\lambda_k'$$

$$E_k = -J_I S\lambda_k'$$

H_0 is a constant term, the coefficients λ_k and λ_k' depend on the crystallographic structure of the magnetic layer. $n^{//}$ represents the number of nearest-neighbors sites in the same atomic plane, while $n_S^{\perp}$ and $n_V^{\perp}$ are the numbers of surface and volume nearest-neighbors in the adjacent plane in the same magnetic layer, respectively. For a given site in the surface plane of the magnetic layer, $n^{\updownarrow}$ represents the number of the nearest neighbors sites in the adjacent layer across the nonmagnetic layer. For fcc (111) ($n^{//} = 6$, $n_S^{\perp} = 3$ and $n_V^{\perp} = 6$) with the lattice constant a and in the case where the nonmagnetic layer does not disturb the succession order of the magnetic atomic planes ($n^{\updownarrow} = 3$):

$$\begin{aligned} \lambda_k &= 4\cos\left(ak_x\frac{\sqrt{6}}{4}\right)\cos\left(ak_y\frac{\sqrt{2}}{4}\right) + 2\cos\left(ak_y\frac{\sqrt{2}}{2}\right) \\ \lambda_k' &= 4\cos\left(ak_x\frac{\sqrt{6}}{12}\right)\cos\left(ak_y\frac{\sqrt{2}}{4}\right) + 2\cos\left(ak_x\frac{\sqrt{6}}{6}\right) \end{aligned} \quad (8)$$

The motion equation is given by:

$$\begin{cases} i\dfrac{\partial b_{\mathbf{k}\alpha\mu}}{\partial t} = \left[b_{\mathbf{k}\alpha\mu}, H\right] \\ i\dfrac{\partial b^{+}_{\mathbf{k}\alpha\mu}}{\partial t} = \left[b^{+}_{-\mathbf{k}\alpha\mu}, H\right] \end{cases} \tag{9}$$

The spin system is characterized by $2nq \times 2nq$ equations, then the resulting secular equation:

$$\begin{cases} \left(C_k + B_k + \omega_{k\alpha\mu}\right) b_{k\alpha\mu} + D_k b_{k\alpha'\mu} + E_k b_{k\alpha'\mu'} + 2Ab^{+}_{-k\alpha\mu} = 0 \\ 2Ab_{k\alpha\mu} + D_k b^{+}_{-k\alpha'\mu} + E_k b^{+}_{-k\alpha'\mu'} + \left(C_k + B_k - \omega_{k\alpha\mu}\right) b^{+}_{-k\alpha\mu} = 0 \end{cases} \tag{10}$$

The secular equation that results from the Heisenberg equation of motion is given by $2n \times 2n$ matrix:

$$W^{(2nq\times 2nq)} = \begin{pmatrix} U^{(nq\times nq)} & V^{(nq\times nq)} \\ \\ -\ V^{(nq\times nq)} & -\ U^{(nq\times nq)} \end{pmatrix}, \quad U^{(nq\times nq)} = \begin{pmatrix} U_1^{(n\times n)} & U_2^{(n\times n)} & & & \\ U_3^{(n\times n)} & U_1^{(n\times n)} & U_2^{(n\times n)} & & \\ & \cdot & \cdot & \cdot & \\ & & \cdot & \cdot & \cdot \\ & & & \cdot & \cdot & \cdot \\ & & & U_3^{(n\times n)} & U_1^{(n\times n)} & U_2^{(n\times n)} \\ & & & & U_3^{(n\times n)} & U_1^{(n\times n)} \end{pmatrix},$$

$$V^{(nq\times nq)} = \begin{pmatrix} V^{(n\times n)} & & & \\ & \cdot & & \\ & & \cdot & \\ & & & \cdot \\ & & & & V^{(n\times n)} \end{pmatrix}, \quad U_1^{(n\times n)} = \begin{pmatrix} B_k & D_k & & & \\ D_k & C_k & D_k & & \\ & \cdot & \cdot & \cdot & \\ & & \cdot & \cdot & \cdot \\ & & & \cdot & \cdot & \cdot \\ & & & D_k & C_k & D_k \\ & & & & D_k & B_k \end{pmatrix}, \quad U_2^{(n\times n)} = \begin{pmatrix} 0 & \cdot & \cdot & \cdot & 0 \\ \cdot & \cdot & & & \cdot \\ \cdot & & \cdot & & \cdot \\ \cdot & & & \cdot & \cdot \\ 0 & & & \cdot & \cdot \\ E_k & 0 & \cdot & \cdot & \cdot & 0 \end{pmatrix},$$

$$U_3^{(n\times n)} = \begin{pmatrix} 0 & \cdot & \cdot & \cdot & 0 & E_k \\ \cdot & \cdot & & & & 0 \\ \cdot & & \cdot & & & \cdot \\ \cdot & & & \cdot & & \cdot \\ \cdot & & & & \cdot & \cdot \\ 0 & & \cdot & \cdot & \cdot & 0 \end{pmatrix}, \quad V^{(n\times n)} = \begin{pmatrix} 2A & & & & & \\ & 0 & & & & \\ & & \cdot & & & \\ & & & \cdot & & \\ & & & & \cdot & \\ & & & & & 0 & \\ & & & & & & 2A \end{pmatrix}$$

We consider the $n \times q$ positive ones which correspond to the $n \times q$ magnon excitation branches $\omega_k^r \left(r = 1, 2, \cdots n \times q\right)$. These branches can be classified into n groups of q quasi-degenerate components in the usual case where J_I remain sufficiently small compared to the effective intralayer exchange strength (**Figure 4**).

The reduced magnetization versus temperature is computed numerically from:

$$m\left(T\right) = 1 - \frac{1}{N_k nqS} \sum_{k,r} \frac{1}{\exp\left(\dfrac{\omega_k^r}{k_B T}\right) - 1} \tag{12}$$

The coefficient N_k indicates the number of k points taken in the first Brillouin zone.

Using Equation (12), satisfactory fits were obtained for the *M* (*T*) data for all of the Ni/Cu multilayer films. The *M* (*T*) theory curves obtained from the fits for all films are shown in **Figure 1**, well matching the experimental data points. Taken $S = 0.3$, and $D^{\perp} = 0$ K, the values of J_b and J_S obtained from the fits are listed in **Table 1**.

The derived bulk exchange interaction constants all consistently fall in the range expected for the bulk exchange interaction of Ni [3,7,13]. The interlayer coupling strength depend on the Ni thickness in the structure, the saturation magnetization (M_S) of the layers and their respective fields ($J_I = \dfrac{M_S H_S t_{Ni}}{4}$, where H_S is the saturation field) [14]. Compared to the bulk exchange interaction coupling, however, the interlayer coupling is con-

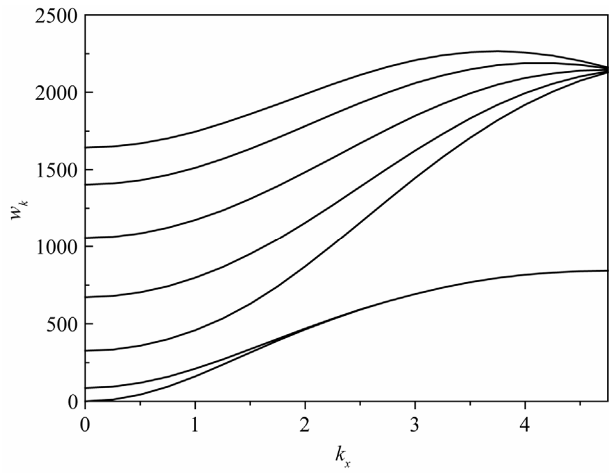

Figure 4. Spin-wave excitation spectrum ω_k. k_x ($k_y = k_x\sqrt{2}$) for fcc (111) ferromagnetic multilayer **with t_{Ni} = 14 Å, S = 0.3, J_S = 77 K, J_b = 240 K, J_I = 0 K, $D^{//}$ = 0.03K, $D^{\perp}$ =0K; in the case: J_b/J_S = 3.**

Table 1. The fitting results from Equation (12) for Ni/Cu. J_b is the bulk exchange interaction between neighboring Ni atoms and J_S is the surface exchange interaction.

t_{Ni} (Å)	J_S/k_b (K)	J_b/k_b (K)	$D^{//}$ (K)
14	70	240	0.03
25	75	240	0.05
50	80	250	0.1

siderably weak. Nonetheless, its effect on the magnetic properties is rather significant. The Ni layers are coupled together by an interlayer exchange coupling, and that the spin waves extend across the whole multilayer sample. The propagation of spin waves through the Cu layers implies the existence of spin polarization within the Cu.

4. Conclusions

The temperature dependence of the magnetization has been investigated for Ni/Cu multilayer. It is seen that the spin-wave constant is much larger than the value found for bulk Ni. A simple model has allowed us to obtain numerical estimates for the exchange interactions and the interlayer coupling strength for Ni/Cu multilayer. Evidence of spin polarization of the nonmagnetic layers, related to the interlayer coupling, was obtained.

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