Exchange interactions in Fe/Pt multilayers

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Available online 28 June 2005

Abstract

The magnetization of Fe/Pt multilayers has been measured as a function of temperature. A bulk-like \( T^{3/2} \) temperature dependence of the magnetization is observed for all multilayers in the temperature range 5–300 K. The spin wave constant \( B \) is found to decrease inversely with \( t_{\text{Fe}} \). A simple theoretical model has been used to explain the temperature dependence of the magnetization and the approximate values for the bulk exchange interaction \( J_b \) and surface exchange interaction \( J_s \) for various Fe layer thicknesses been obtained.

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Keywords: Fe/Pt multilayers; Magnetization; Spin wave excitations; Exchange interactions

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1. Introduction

The magnetic behavior of two-dimensional systems has been studied both theoretically and experimentally by many authors because it may help to understand fundamental properties of magnetically ordered systems. One common approach is to study the spontaneous magnetization, its temperature dependence and the Curie temperature, $T_C$, of ferromagnetic thin films. Experimentally, a drastic change of the magnetic behavior is found for films thinner than a critical thickness, which varies from author to author [1–5]. The basic problem is whether these observations describe intrinsic properties of two-dimensional ferromagnets or are mainly produced by structural imperfections (contamination, diffusion, island growth).

Recently, Pinettes and Lacroix examine the influence of anisotropy on the thickness dependence of spin wave excitation spectra and calculate the thermal variation of the magnetization as a function of film thickness [6]. It has been shown that the surface anisotropy strongly affects the thickness dependence of the magnetization. In the case of a multilayer the thermal variation of magnetization and the Curie temperature is also affected by the interlayer exchange coupling [7,8]. Furthermore, interdiffusion causing graded interfaces and disorder resulting in a distribution of exchange interactions can play an important role. Especially when the magnetic layer thickness is in the monolayer regime, the latter two factors might dominate the temperature dependence.

In this paper we present our studies on Fe/Pt multilayers prepared by the RF sputtering method. Assuming a Heisenberg framework, we propose a simple theoretical model, including both intralayer and interlayer exchange interactions. The exchange interactions strengths are adjusted directly by fitting the calculated magnetization to the measured one.

2. Experimental

The multilayers were deposited onto water-cooled glass substrates by RF diode sputtering. The chamber was first evacuated to a pressure of $1–2 \times 10^{-7}$ Torr using a turbomolecular pump. Argon of 5 N purity was used as the sputter gas and its pressure was kept constant at $6 \times 10^{-3}$ Torr. The RF power density was 2.1 W/cm$^2$. The thickness was measured in situ using a pre-calibrated quartz monitor. All the samples were grown on Pt buffer layers 100 Å thick. The magnetic layer thickness $t_{Fe}$ was varied in the range 8.4–57 Å and that of the Pt layer $t_{Pt}$ was fixed at 18 Å. X-ray diffraction profiles, taken in reflection geometry at both low ($2\theta < 10^\circ$) and high ($35^\circ < 2\theta < 60^\circ$) scattering angle, confirmed the [110]. The magnetization $M$ was measured using a vibrating sample magnetometer (VSM).

3. Results and discussion

Fig. 1 shows the temperature dependence of $M$ for several values of $t_{Fe}$ thicknesses. It can be noticed that $T_C$ decreases when $t_{Fe}$ decreases.
The low-temperature magnetization was studied in detail for a few samples. For three-dimensional magnetic films, the magnetization has a $T^{3/2}$ dependence due to classical spin-wave excitations. In such cases, according to spin-wave theory, the temperature dependence should follow the relation

$$\frac{M(5\,\text{K}) - M(T)}{M(5\,\text{K})} = BT^{3/2}.$$  

In all cases this behavior is observed for temperatures as high as $T_C/3$. The spin-wave constant $B$ decreases from $48 \times 10^{-6}\,\text{K}^{-3/2}$ for $t_{\text{Fe}} = 8.4\,\text{Å}$ to $7.3 \times 10^{-6}\,\text{K}^{-3/2}$ for $t_{\text{Fe}} = 57\,\text{Å}$. These values are much larger than the value of $5 \times 10^{-6}\,\text{K}^{-3/2}$ found for bulk Fe. The $B$ vs. $1/t_{\text{Fe}}$ is plotted for the samples with $8.4 \leq t_{\text{Fe}} \leq 57\,\text{Å}$ in Fig. 2.

It is seen that the experimental points align well in a straight line. The values extrapolated to $1/t_{\text{Fe}} = 0$ are in good agreement with those found for the bulk Fe. It was observed that the parameters $B$ in Eq. (1) depend on $t_{\text{Fe}}$ according to

$$B(t_{\text{Fe}}) = B_\infty + B_s/t_{\text{Fe}},$$

where $B_\infty$ is the bulk spin wave parameter of Fe and $B_s$ the surface $B$ value. The interface anisotropy strongly affects the thickness dependence of the magnetization. The linear relation between the spin wave parameter $B$ and the reciprocal of the
magnetic film thickness was reported also by Gradmann and co-workers [9, 10] for Fe(1 1 0) films on W(1 1 0).

The apparent universality of Bloch’s $T^{3/2}$ law for the temperature dependence of the spontaneous magnetization, and of generalization thereof, is considered by Krey [11]. It is argued that in the derivation one should not only consider the exchange interaction between the spin, but also the other interactions between them, leading to elliptical spin precession and deviations from the parabolic dispersion of magnons. Also interaction effects are important to explain the apparent universality of generalized Bloch law exponents $n$, defined by $M(T) = M(0) - \text{const.} T^n$, valid in a wide temperature range, and for dimensionality $d = 1, 2$, and 3. However, due to interactions, the Bloch exponent $n$ depends not only on the dimensionality $d$ of the system, but also on the spin quantum number $S$ of the system [11].

To describe the experiment results we suppose that the multilayer $(X_n/Y_m)_q$ is formed by an alternate deposition of a magnetic layer (X) and non-magnetic one (Y). The multilayer is characterized by the number ($q$) of pair (X/Y), the number ($n$) of atomic planes in the magnetic layer $\mu$ and the number ($m$) of atomic planes in the non-magnetic layer. We chose the lattice unit vectors ($e_x$, $e_y$, $e_z$) so that $e_z$ is perpendicular to the atomic planes. We note by $S_{iz\mu}$ the spin operator of the atom $i (i = 1, 2, \ldots, N)$ in the plane $\alpha (\alpha = 1, 2, \ldots, n)$ of the magnetic layer $\mu (\mu = 1, 2, \ldots, q)$. Further, we suppose that the multilayer is characterized by a rigid lattice and by perfectly sharp layer interfaces without structural imperfections (contamination, diffusion, island growth, etc.). In this framework the system

![Fig. 2. $t_{Fe}^{-1}$ dependence of $B$.](image)
Hamiltonian is given by
\[
H = - J_b \left[ \sum_{\langle \mu, jz_d \rangle} S_{i\mu} S_{jz_d} + \sum_{\langle \mu, jz_d' \mu' \rangle} S_{i\mu} S_{jz_d'} \right] 
- J_s \sum_{\langle \mu, jz_s \rangle} S_{i\mu} S_{jz_s} - J_1 \sum_{\langle \mu, jz_s' \mu' \rangle} S_{i\mu} S_{jz_s' \mu'} ,
\]
(3)
\[
H \text{ describes the exchange interactions in the same magnetic layer (bulk and surface) as well as the exchange interactions between adjacent magnetic layers. } J_b \text{ and } J_s \text{ are the bulk and surface exchange interactions, } J_1 \text{ is the strength of the interlayer coupling restricted to the surface layers. Further we denote by } \sum_{\Xi} \text{ the summation on the sites of the bulk layer planes (} \Xi = b, \text{ surface layer planes (} \Xi = s) \text{ or the surfaces planes coupled via the non-magnetic layer (} \Xi = I). The symbol } \langle \rangle \text{ denotes the pairs of nearest-neighbor atoms or adjacent magnetic planes.}
\]
In the Holstein–Primakoff formulation [12], the creation and annihilation operators (\(a_{i\mu}^+\) and \(a_{i\mu}\)) for each atomic spin are related to the spin operators by
\[
S_{i\mu}^X + iS_{i\mu}^Y = (2S)^{1/2} f_{i\mu}(2S) a_{i\mu} \quad \text{and} \quad S_{i\mu}^X - iS_{i\mu}^Y = (2S)^{1/2} a_{i\mu}^+ f_{i\mu}(2S) .
\]
(4)
In the framework 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 (\(T < T_C/3\)) [13,14]. So, the value of \(f_{i\mu}(2S)\) is fixed to 1. We pass from the atomic variables to the magnon variables after a two-dimensional Fourier transformation. We show that
\[
H = H_0 + \sum_{k,\langle \mu, \phi_{z}\rangle} A_k b_{kz\mu}^+ b_{kz\mu} + \sum_{k,\langle \mu, \phi_{z}\rangle} B_k b_{kz\mu}^+ b_{kz\mu}
+ \sum_{k,\langle \mu, \phi_{z}\rangle} C_k b_{kz\mu}^+ b_{kz\mu} + \sum_{k,\langle \mu, \phi_{z}\rangle} D_k b_{kz\mu}^+ b_{kz\mu} ,
\]
(5)
where
\[
A_k = (J_s(2n^\parallel - (\lambda^\parallel_k + \lambda^-_k)) + 2J_b n^\perp) S + 2J_1 n^\parallel S ,
B_k = (4n^\parallel + 2n^\perp - (\lambda^\parallel_k + \lambda^-_k)) J_b S ,
C_k = -J_b S \lambda^\parallel_k ,
D_k = -J_1 S \lambda^\parallel_k .
\]
(6)
\(H_0\) is a constant term, and the coefficients \(\lambda^\parallel_k, \lambda^-_k, \lambda^\parallel_k'\) and \(\lambda^-_k\) depend on the crystallographic structure of the magnetic layer. \(n^\parallel\) represent the number of nearest-neighbor sites in the same atomic plane, while \(n^\perp (n^\parallel)\) is the number of nearest neighbors in the adjacent plane in the same (adjacent) magnetic layer. For BCC(1 1 0) (\(n^\parallel = 4\) and \(n^\perp = 2\)) with the lattice constant \(a\) and in the case where the non-magnetic layer does not disturb the succession order of the magnetic atomic
planes \((n^\dagger = 2)\),

\[
\lambda^+_k = \lambda^-_k = 4 \cos(ak_x \sqrt{2}/2) \cos(ak_y/2),
\]

\[
\lambda'_k = \lambda''_k = 4 \cos(ak_y/2).
\]

(7)

The spin system is characterized by \(2nq \times 2nq\) equations; then the resulting secular equation is given by a \((2nq \times 2nq)\) matrix:

\[
W^{(2nq\times2nq)} = \begin{pmatrix}
U^{(nq\times nq)} & -U^{(nq\times nq)} \\
U_1^{(n\times n)} & U_2^{(n\times n)} & U_3^{(n\times n)} & \ldots & U_{nq}^{(n\times n)} \\
U_1^{(n\times n)} & U_2^{(n\times n)} & U_3^{(n\times n)} & \ldots & U_{nq}^{(n\times n)} \\
\vdots & \vdots & \vdots & \ddots & \vdots \\
U_1^{(n\times n)} & U_2^{(n\times n)} & U_3^{(n\times n)} & \ldots & U_{nq}^{(n\times n)}
\end{pmatrix}
\]

where \(U^{(nq\times nq)} = \)

\[
\begin{pmatrix}
B_k & D_k & D_k & \ldots & D_k \\
D_k & C_k & D_k & \ldots & \ldots \\
\vdots & \vdots & \vdots & \ddots & \vdots \\
D_k & C_k & D_k & \ldots & B_k \\
0 & \cdots & 0 \\
\cdots & \cdots & \cdots & \cdots & \cdots \\
0 & \cdots & 0 & \cdots & 0 \\
E_k & 0 & \cdots & 0 & 0
\end{pmatrix}
\]

\( \text{and } U_3^{(n\times n)} = \)

\[
\begin{pmatrix}
0 & \cdots & 0 & E_k \\
\cdots & \cdots & \cdots & 0 \\
\cdots & \cdots & \cdots & \cdots \\
0 & \cdots & 0 & 0
\end{pmatrix}
\]

(8)

Among the \(2(n \times q)\) eigenvalues of the matrix \(W^{(2nq\times2nq)}\), we consider the \(n \times q\) positive ones which correspond to the \(n \times q\) magnon excitation branches \(\omega_k^r (r = 1, 2, \ldots, n \times q)\). These branches can be classified into \(n\) groups of \(q\)
quasi-degenerate components in the usual case where $J_1$ remain sufficiently small compared to the effective intralayer exchange strength (Fig. 3).

The reduced magnetization $m(T)$ vs. temperature is computed numerically from

$$m(T) = 1 - \frac{1}{N_k n q S} \sum_{k,r} \frac{1}{\exp(\omega_k^r/k_B T) - 1}.$$  \hspace{1cm} (9)

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

Using Eq. (9), satisfactory fits were obtained for the $m(T)$ data for all of the Fe/Pt multilayer films. The $m(T)$ theory curves obtained from the fits are shown in Fig. 1, well matching the experimental data points. The values of $J_b$, $J_s$, $J_1$ obtained from the fits are listed in Table 1 for all films (taken $S = 1$).

![Fig. 3. Spin wave excitation spectrum vs. $k_x (k_y = k_x \sqrt{2})$ for BCC(110) ferromagnetic multilayer with $q = 20; n = 3; J_s = 40$ K; $J_b = 100$ K; $J_1 = 5$ K; $S = 1$.](image)

Table 1
The fitting results from Eq. (9) for Fe($t_{Fe}$)/Pt($t_{Pt} = 18$ Å)

<table>
<thead>
<tr>
<th>$t_{Fe}$ (Å)</th>
<th>$J_b/k_B$ (K)</th>
<th>$J_s/k_B$ (K)</th>
<th>$J_1/k_B$ (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>8.4</td>
<td>101</td>
<td>25</td>
<td>0.9</td>
</tr>
<tr>
<td>24</td>
<td>94</td>
<td>26</td>
<td>0.3</td>
</tr>
<tr>
<td>57</td>
<td>146</td>
<td>68</td>
<td>0.8</td>
</tr>
</tbody>
</table>

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The derived bulk exchange interaction constants all consistently fall in the range expected for the exchange interaction of bulk Fe [15,16]. In the literature, the order of the interlayer coupling strengths is diverse [17–19]. For this parameter, our results remain in the same order but lesser than that reported by Gutierrez et al. in Fe/Ag multilayers [20].

4. Conclusions

In conclusion, the temperature dependence of the magnetization of Fe/Pt multilayers has been investigated for various Fe layer thicknesses. The thermal variation of the magnetization in ferromagnetic multilayer films is calculated using spin-wave theory. This simple model has allowed us to obtain numerical estimates for the bulk exchange interaction $J_b$, surface exchange interaction $J_s$ and the interlayer coupling strength $J_I$ at various Fe layer thicknesses.

References