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Spin-wave excitations and magnetism of sputtered Fe/Au multilayers

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Abstract. The spin-wave excitations and the magnetism of Fe/Au multilayers with different Fe thicknesses (t_{Fe}) grown by RF sputtering were investigated. The temperature dependence of spontaneous magnetization is well described by a $T^{3/2}$ law in all multilayers in the temperature range of 5–300 K. Spin-wave theory has been used to explain the temperature dependence of the spontaneous magnetization and the approximate values for the exchange interactions for various t_{Fe} were obtained. The spin-wave constant B was found to increase linearly with the inverse in the Fe thickness ($1/t_{Fe}$). Using the ferromagnetic resonance technique, the change of the anisotropy field H_K as a function of $1/t_{Fe}$ was deduced. The spatial distributions of the discrete spin-wave modes were calculated. All the extracted results were in agreement with those determined experimentally and found in the literature.

Keywords. Fe/Au multilayers; magnetization; spin-wave excitations; exchange interactions.

1. Introduction

Magnetism in ultrathin films has been intensively studied due to the peculiar magnetic properties existing in such systems, with a high potential for spintronic devices [1]. Magnetic multilayers exhibit interesting properties, such as giant magnetoresistance, interlayer exchange coupling and enhancement of magnetic moment of ferromagnetic atoms [2–4]. The period, number of layers and the relative thicknesses of layers affect significantly the multilayer properties, which are in turn sensitive to the microstructure [5–7]. The properties of multilayers are mostly governed by the surface state and hence, the interface plays a key role. The discovery of coupled magnetic behaviour between layer components in different magnetic multilayers has led to an increased interest in two-dimensional systems. In most ferromagnetic materials and below the T_C, the temperature dependence of the spontaneous magnetization $M(T)$ is well described by Bloch's law resulting from the linearized spin-wave theory for bulk ferromagnets [8]. However, for two-dimensional ferromagnets, simulations within the framework of spin-wave theory do not usually predict a $T^{3/2}$ law for the $M(T)$ curve. In contrast, empirical data have verified the validity of Bloch's law for ultrathin ferromagnetic films in many cases [7]. A possible explanation for this unexpected fact was explained by Mathon and Ahmed [9] who predicted an 'effective $T^{3/2}$ law' to be applicable in a certain temperature range for two-dimensional systems.

On the other hand, the spin-wave excitations are also important parameters in magnetic systems [10–12], which play a crucial role on their magnetic properties at low temperature and they also describe the time scale for the magnetization dynamics. In this paper, the spin-wave excitations and thermal variation of the spontaneous magnetization in Fe/Au multilayers as a function of Fe thickness were studied and compared qualitatively and quantitatively with experimental results.

2. Experimental

Fe/Au multilayers were prepared by RF-sputtering technique. The base pressure was less than $1–2 \times 10^{-7}$mbar and the working pressure of Ar was 6.6×10^{-3} mbar for deposition. The deposition was carried at room temperature. The thickness was measured *in situ* using a pre-calibrated quartz monitor. The Fe layer thickness (t_{Fe}) was varied from 7 to 72 Å and the Au layer thickness t_{Au} was fixed at 20 Å. The number of bilayers was in the range of 10–20. All the samples were grown on a Au buffer layer of 100 Å thick and covered by a 20 Å thick Au layer to prevent oxidation. In all cases, the first and the last layers were Au. The substrates were floated on glass plates. X-ray diffraction measurements, taken in reflection geometry at both low ($2\theta < 10°$) and high ($30° \leq 2\theta \leq 50°$) scattering angles, confirmed the modulated structure and showed a (110) texture for *bcc* Fe. The *fcc* Au buffer layer is seen to have a (111) texture. Magnetization was measured using a vibrating sample magnetometer with an applied magnetic field up to 2 T.

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3. Results and discussion

The in-plane M–H hysteresis curves present a rectangular shape (data not shown). The coercivity increases slightly from 3.8 to 9.6 Oe with decreasing t_{Fe} from 40 to 15 Å at 300 K, which can be seen by the increase of the interface contribution with decreasing film thickness, which should induce an increase in the random anisotropy for thinner Fe layers. The interlayer exchange coupling strength, J_I can be expressed using the following formula: $J_I = M_S \cdot H_S \cdot t_{Fe}/4$, where M_s is the saturation magnetization and H_S is the saturation field [5]. It is found that J_I increases from 5×10^{-3} to 10^{-2} erg cm^{-2}, when t_{Fe} increases from 15 to 40 Å.

The magnetization was measured at fields up to 2 T, between 5 and 300 K using a vibrating sample magnetometer and it is expressed in terms of total volume of the Fe layers. As shown in figure 1, at 300 K and Fe thicknesses ranging from 24 to 72 Å, the magnetization is found to be equal to 1700 ± 80 emu cm^{-3} but for t_{Fe} smaller than 24 Å, the magnetization increases with the decrease in Fe thickness.

Figure 2 shows the temperature dependence of magnetization for different values of t_{Fe}. It can be noticed that T_C decreases with decreasing t_{Fe}. The decrease of T_C with decreasing t_{Fe} is evident from the change in the slope of the magnetization curve and can be noted by the reduction in the Fe–Fe exchange interaction as a result of finite size effects. The low-temperature magnetization was studied in detail for 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 be written as

$$\frac{M(5\mathrm{K}) - M(\mathrm{T})}{M(5\mathrm{K})} = BT^{3/2}, \tag{1}$$

where B is the spin-wave constant.

As observed in figure 2, in all cases, the M(T) curves are well fitted for temperatures as high as $T_C/3$ (solid lines). It was noted that B parameter can be expressed as a function of Fe thickness using the following formula [13,14]:

$$B(t_{Fe}) = B_{bulk} + \frac{B_{surface}}{t_{Fe}}, \tag{2}$$

where B_{Bulk} is the bulk spin-wave constant of Fe and $B_{surface}$ the B value for the surface contribution. It was found that the interface anisotropy can affect the thickness dependence of the magnetization. The $B(t_{Fe})$ *vs.* $1/t_{Fe}$ curve for samples with Fe thicknesses ranging from 10 to 72 Å is reported in figure 3. It was found that B decreases from 21×10^{-6} to 7.2×10^{-6} K$^{-3/2}$ with increasing t_{Fe} from 10 to 72 Å, respectively. The experimental points fit reasonably with the straight line. The extrapolation of the linear fit gives the value of the bulk spin-wave constant which is equal to $B_{bulk} = 4.9 \times 10^{-6}$ K$^{-3/2}$,

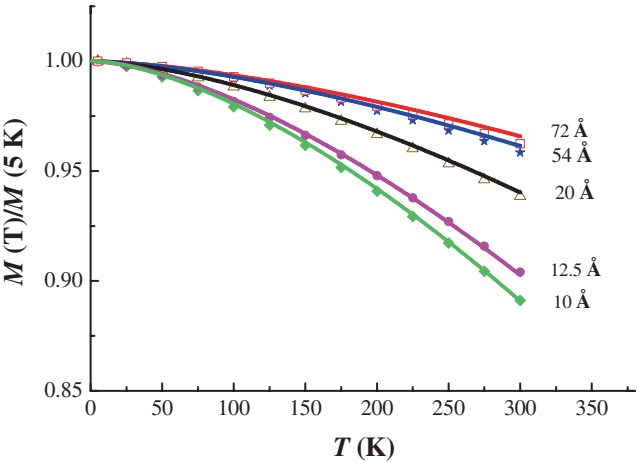

Figure 2. Measured (symbols) and calculated (continuous line) temperature dependence of the normalized magnetization of Fe/Au multilayers for different Fe thicknesses.

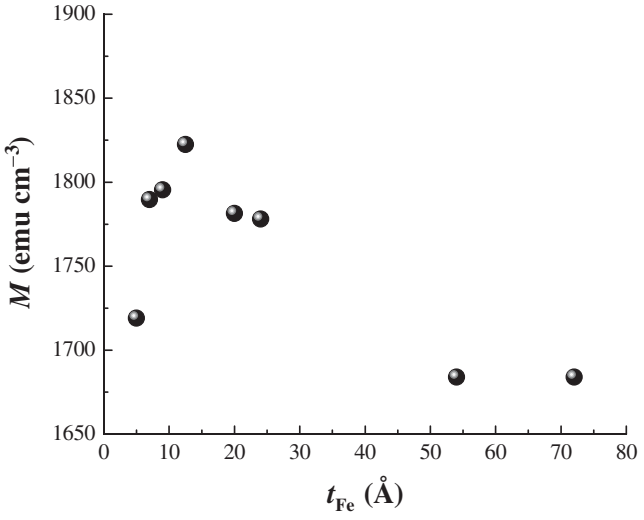

Figure 1. Magnetization *vs.* Fe thickness recorded at 300 K for Fe/Au multilayers, the Au thickness was fixed at 20 Å.

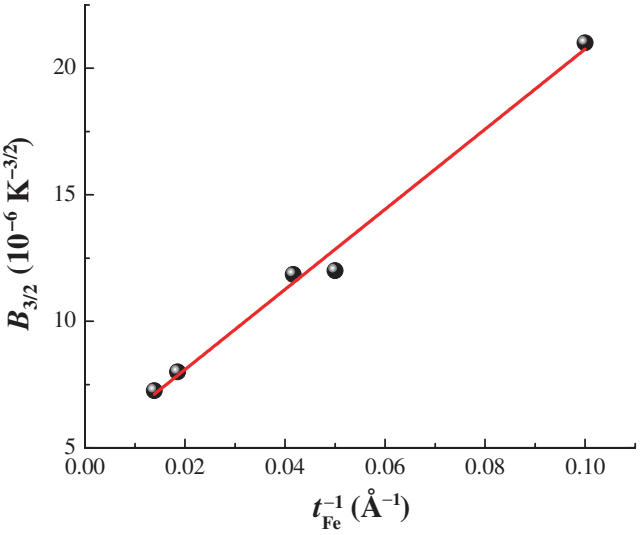

Figure 3. The spin-wave constant (B) as a function of t_{Fe}^{-1}.

and it is in good agreement with the value obtained for macroscopic-sized samples of bulk *bcc* Fe [15]. From the slopes of the straight line, the B value on surface, B_{surface} is deduced to be 1.6×10^{-4} Å $\text{K}^{-3/2}$.

To obtain the g-factor ($g = 2.05 \pm 0.03$) and effective field magnetization, $4\pi M_{\text{eff}}$, ferromagnetic resonance measurements with frequencies ranging from 25.09 to 35.73 GHz with the field applied both parallel and perpendicular to the film plane were carried out. The uniaxial anisotropy field, H_{U}, was determined by knowing M_{S} from the magnetic measurements ($H_{\text{U}} = 4\pi M_{\text{S}}$).

According to previous studies [3,13], an interface magnetic anisotropy of multilayers can be seen through the magnetic layer thickness dependence of the perpendicular anisotropy field. If the interface anisotropy mainly increases the first-order anisotropy energy K_1, then, the perpendicular anisotropy field (excluding the demagnetization term) can be expressed as

$$H_{\text{K}} = 4\pi M_{\text{S}} - 4\pi M_{\text{eff}} = H_{\text{U}} + 2H_{\text{SA}}/t_{\text{Fe}}, \tag{3}$$

where H_{U} is the volume anisotropy field and H_{SA} the surface contribution to the anisotropy field. It was noticed that the perpendicular anisotropy field can also be expressed as $H_{\text{K}} = 2K_1/M_{\text{S}}$. H_{U} and H_{SA} can also be written as $H_{\text{U}} = 2K_{\text{U}}/M_{\text{S}}$ and $H_{\text{SA}} = 2K_{\text{S}}/M_{\text{S}}$.

Figure 4 shows the plot of H_{K} as a function of $1/t_{\text{Fe}}$. A linear variation of H_{K} *vs.* $1/t_{\text{Fe}}$ can be observed. From the slope of the straight line, the value of the interface anisotropy constant K_{S} is deduced to be 0.38 erg cm^{-2} at 300 K. The positive value of K_{S} indicates that demagnetization energy is still dominating, which means that the perpendicular magnetization is not stabilized in these multilayers.

Theoretical calculations were performed using a model for spin waves in ferromagnetic/nonmagnetic multilayers as described in reference [14]. The basic characteristics may be summarized as follows: the multilayer $(\text{Fe}_n/\text{Au}_m)_q$ is

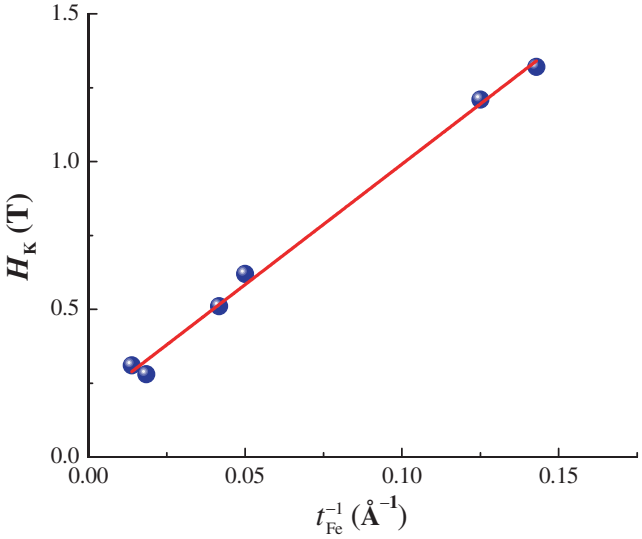

Figure 4. Change of H_{K} as a function of t_{Fe}^{-1} for Fe/Au multilayers.

supported to be formed by an alternate deposition of a magnetic layer (Fe) and nonmagnetic layer (Au). The multilayer is characterized by the number (q) of bi-layers (Fe/Au). The number of atomic planes in the magnetic and nonmagnetic layers are denoted n and m, respectively.

The lattice unit vectors ($\vec{e}_{\text{X}}, \vec{e}_{\text{Y}}, \vec{e}_{\text{Z}}$) was chosen, so that $\vec{e}_{\text{Z}}$ is perpendicular to the atomic planes. $\vec{S}_{i\alpha\mu}$ the spin operator of the atom i ($i = 1, 2, \ldots, N$) in the plane α ($\alpha = 1, 2, \ldots, n$) of the magnetic layer μ ($\mu = 1, 2, \ldots, q$) was observed. Furthermore, it was assumed that the multilayer is characterized by a rigid lattice and perfect sharp layer interfaces without structural imperfections (contamination, diffusion, island growth, etc.).

The full Hamiltonian system can be expressed as a summation of two terms

$$H = H_{\text{e}} + H_{\text{s}}. \tag{4}$$

H_{e} describes the exchange interactions in the same magnetic layer (bulk and surface) as well as the exchange interactions between adjacent magnetic layers which can be written as

$$\begin{aligned} H_{\text{e}} &= -J_{\text{b}} \left[\sum_{\langle i\alpha\mu j\alpha\mu\rangle}^{b} S_{i\alpha\mu} S_{j\alpha\mu} + \sum_{\langle i\alpha\mu j\alpha'\mu\rangle} S_{i\alpha\mu} S_{j\alpha'\mu} \right] \\ &\quad - J_{\text{s}} \sum_{\langle i\alpha\mu j\alpha\mu\rangle}^{s} S_{i\alpha\mu} S_{j\alpha\mu} - J_{\text{I}} \sum_{\langle i\alpha\mu j\alpha''\mu''\rangle}^{I} S_{i\alpha\mu} S_{j\alpha''\mu''}. \end{aligned} \tag{5}$$

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 non-magnetic layer. $\sum^{b}$ and $\sum^{s}$ are the summations on the sites of bulk and surface layer planes. $\sum^{I}$ is the contribution for the surfaces planes coupled via the nonmagnetic layer. The summation with $\langle\rangle$ denotes the pairs of nearest-neighbour atoms within the same plane or adjacent magnetic planes.

The contribution of the surface anisotropy is estimated by using the following formula:

$$H_{\text{s}} = D^{\perp} \sum_{i\alpha\mu}^{s} (S_{i\alpha\mu}^{\text{Z}})^2 + D^{\parallel} \sum_{i\alpha\mu}^{s} \left((S_{i\alpha\mu}^{\text{X}})^2 - (S_{i\alpha\mu}^{\text{Y}})^2 \right). \tag{6}$$

$D^{\perp}$ and $D^{\parallel}$ are the surface anisotropy parameters for the uniaxial out-of-plane and in-plane components, respectively.

In the Holstein–Primakoff formulation [16], 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

$$\begin{aligned} S_{i\alpha\mu}^{\text{X}} + i S_{i\alpha\mu}^{\text{Y}} &= (2S)^{1/2} f_{i\alpha\mu}(2S)\, a_{i\alpha\mu}, \\ S_{i\alpha\mu}^{\text{X}} - i S_{i\alpha\mu}^{\text{Y}} &= (2S)^{1/2} a_{i\alpha\mu}^{+} f_{i\alpha\mu}(2S). \end{aligned} \tag{7}$$

In the framework of non-interacting spin-wave theory, the linear approximation of the Holstein–Primakoff method is sufficient to describe the magnetic behaviour and the correction terms are quite-small at low temperatures ($T < T_{\text{C}}/3$). As a result, the value of $f_{i\alpha\mu}(2S)$ can be fixed to 1 [17].

The terms $a_{i\alpha\mu}$, $a^{+}_{i\alpha\mu}$ were replaced by the magnon variables ($b_{k\alpha\mu}$, $b^{+}_{k\alpha\mu}$) after a two-dimensional Fourier transformation and thus, the following formula was obtained:

$$\begin{aligned} H &= H_0 + A\sum_{\vec{k}\alpha\mu}^{S}\left(b_{\vec{k}\alpha\mu}b_{-\vec{k}\alpha\mu} + b^{+}_{\vec{k}\alpha\mu}b^{+}_{-\vec{k}\alpha\mu}\right) \\ &\quad + \sum_{\vec{k}\alpha\mu}^{S} B_k b^{+}_{\vec{k}\alpha\mu}b_{\vec{k}\alpha\mu} + \sum_{\vec{k}\alpha\mu}^{b} C_k b^{+}_{\vec{k}\alpha\mu}b_{\vec{k}\alpha\mu} \\ &\quad + \sum_{\vec{k}(\alpha\mu,\alpha'\mu)} D_k b^{+}_{\vec{k}\alpha\mu}b_{\vec{k}\alpha'\mu} \\ &\quad + \sum_{\vec{k}(\alpha\mu,\alpha''\mu'')}^{I} E_k b^{+}_{\vec{k}\alpha\mu}b_{\vec{k}\alpha''\mu''}, \end{aligned} \tag{8}$$

where

$$\begin{aligned} A &= \frac{S}{2}\left(D^{\perp} - D^{||}\right), \\ B_k &= 2S\left(J_s(n^{||} - \lambda_k) + J_b n^{\perp} + J_I n^{\updownarrow}\right) \\ &\quad + S(3D^{||} + D^{\perp}), \\ C_k &= 2J_b S\left((n^{||} - \lambda_k) + 2n^{\perp}\right), \\ D_k &= -J_b S\lambda'_{\vec{k}}, \\ E_k &= -J_I S\lambda'_{\vec{k}}. \end{aligned} \tag{9}$$

H_0 is a constant term and $D^2_{\mathrm{eff}} = D^2_{\perp} + D^2_{||}$. $D_{\mathrm{eff}}\,(K) = K_S\, a^2/k_B$, where a is the lattice constant and k_B the Boltzmann constant.

The coefficients λ_k and λ'_k depend on the crystallographic structure of the magnetic layer. $n^{||}$ represents the number of nearest-neighbour sites in the same atomic plane, while $n^{\perp}_S$ and $n^{\perp}_V$ are the numbers of surface and volume nearest neighbours 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 nearest-neighbour sites in the adjacent layer across the nonmagnetic layer. For *bcc* (110), $n^{||} = 4$ and $n^{\perp} = 2$, with the lattice constant a and in the case where the nonmagnetic layer does not disturb the succession order of the magnetic atomic planes ($n^{\updownarrow} = 2$):

$$\lambda_k = 4\cos(ak_x\sqrt{2}/2)\cos(ak_y/2) \text{ and } \lambda'_k = 4\cos(ak_y/2). \tag{10}$$

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

$$\begin{cases} \left(C_k + B_k + \omega_{k\alpha\mu}\right) b_{k\alpha\mu} + D_k b_{k\alpha'\mu} \\ \quad + E_k b_{k\alpha'\mu'} + 2A\, b^{+}_{-k\alpha\mu} = 0, \\ 2A\, b_{k\alpha\mu} + D_k b^{+}_{-k\alpha'\mu} + E_k b^{+}_{-k\alpha'\mu'} \\ \quad + \left(C_k + B_k - \omega_{k\alpha\mu}\right) b^{+}_{-k\alpha\mu} = 0. \end{cases} \tag{11}$$

We consider the $n \times q$ as positive ones which correspond to the $n \times q$ magnon excitation branches ω^r_k ($r = 1, 2, \ldots n \times q$). As can be seen in figure 5, these branches can be classified into n groups of q quasi-degenerate components

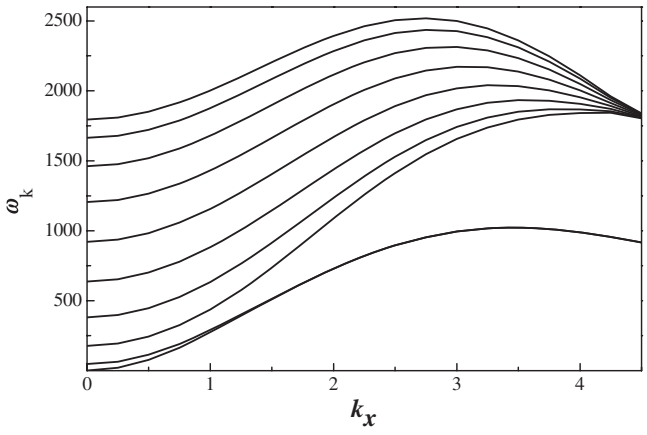

Figure 5. Spin-wave excitation spectrum as a function of k_x ($k_y = k_x\sqrt{2}$) for Fe/Au multilayers, in the case of: $q = 1$, $n = 10$; $S = 1.15$; $J_b = 100$ K, $J_S = 50$ K; $D^{||} = 0.9$ K; $D^{\perp} = -2$ K and $J_I = 10^{-2}$ K.

as expected, in the usual case where J_I remains sufficiently small compared to the effective interlayer exchange strength. The reduced magnetization *vs.* temperature is computed numerically from

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

The coefficient N_k indicates the number of k points taken in the first Brillouin zone. In equation (12), the zero-point fluctuation effects have not been taken into account.

Taken $S = 1.15$, $D^{||} = 0.9$ K and $D^{\perp} = -2$ K (0.38 erg cm^{-2}), the values of J_b and J_S are found to be equal to 90 ± 5 and 45 ± 5 K, respectively, for all multilayers. The deduced bulk exchange interaction constants are consistent with the expected range for the Fe bulk exchange interaction [18]. The interlayer coupling strength is found to change from 10^{-3} to 10^{-2} K, with increasing t_{Fe} from 10 to 72 Å, respectively. We note that different values were reported in the literature for the interlayer coupling strengths as found in refs [19–21]. However, our results remain in the same order but a bit smaller than that reported by Gutierrez *et al* [22] in Fe/Ag multilayer. Compared to the bulk exchange interaction coupling, however, the interlayer coupling is considerably weaker and its effect on the magnetic properties is negligible.

4. Conclusion

In conclusion, Fe/Au multilayers were prepared by RF sputtering and their spin-wave excitations and magnetic properties were studied. The thermal variation of the magnetization in our multilayers is modelled using spin-wave theory. A simple model has allowed us to obtain numerical estimates of different fundamental constants. The spatial distributions of the discrete spin-wave modes were also determined. Reasonable agreement with the experimental data was reported.

Finally, this study will be useful to understand the magnetic properties of Fe/Au multilayers.

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