Extending the Snoek's limit of single layer film in $(Co_{96}Zr_4/Cu)_n$ multilayers

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The present work investigates the high frequency characteristics of $(Co_{96}Zr_4/Cu)_n$ multilayers. The results reveal that the Snoek's limit of $(Co_{96}Zr_4/Cu)_n$ multilayers (revised Acher's limit) are much larger (as a fact 1.76 for some samples) than that of single layer {Acher's limit [O. Acher and A. L. Adenot, Phys. Rev. B **62**, 11324 (2000)]} by comparing their static and dynamic magnetism. It is found that the differences of Acher's limit between multilayers and single layers are caused by magnetic interface anisotropy. This work might facilitate search for new materials with high permeability at high frequency. © *2008 American Institute of Physics*. [DOI: 10.1063/1.3003876]

High permeability at high frequency of magnetic materials is a persistent requirement for the ceaseless increase of the working frequency of the data transmission and multiple accesses as well as the reduction of the electromagnetic interference in computer, mobile and blue-tooth devices.[1–3] However, according to work of Snoek,[4] there exist trade-offs between permeability and resonance frequency. That is to say, the higher the resonance frequency f_r, the lower the low frequency permeability μ_s. For a bulk polycrystalline material, the static permeability μ_s and the resonance frequency f_r of most bulk magnets with saturation magnetization M_s and uniaxial magnetocrystalline anisotropic field H_k fit the function[4]

$$L_S = (\mu_s - 1)f_r = \frac{\gamma}{3\pi}4\pi M_s, \tag{1}$$

where $\mu_s - 1 = 2(4\pi M_s)/3H_k$, $f_r = \gamma H_k/2\pi$, γ is the gyromagnetic factor.

This relationship can be extended to Acher's limit in magnetic thin films with uniform in-plane uniaxial magnetic anisotropies[5] as

$$L_A = (\mu_s - 1)f_r^2 = \left(\frac{\gamma}{2\pi}4\pi M_s\right)^2, \tag{2}$$

where $\mu_s - 1 = 4\pi M_s/H_k$, and f_r in these thin films can be written as Kittel equation.[6]

$$f_r = \frac{\gamma}{2\pi}\sqrt{4\pi M_s H_k}. \tag{3}$$

However, some researchers found that the resonance frequency of Permalloy films measured by pulsed inductive microwave magnetometer is larger than that calculated by the Kittel equation.[7–9] The authors explained that this result is caused by surface magnetostriction between the substrate and the thin film. This means that the Acher's limit of Permalloy films can be extended in some single layer films with other interactions except in-plane uniaxial anisotropy. In this work, the $(Co_{96}Zr_4/Cu)_n$ multilayers with in-plane interfaces anisotropy were used to get larger Acher's limit in multilayers.

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The multilayers were prepared by radio frequency (rf) sputtering onto $10\times20\times0.42$ mm^3 (111)-oriented Si substrates attached to a water-cooling system with background pressure lower than 5×10^{-5} Pa. A Cu target, 70 mm in diameter and 3 mm in thickness, was used to deposit Cu layers, and a Co target, same size as Cu target, on which Zr chips were placed in a regular manner, was used to deposit $Co_{96}Zr_4$ layers. Magnetic layers were deposited at an angle of 24° to attain uniaxial anisotropy.[10,11] The composition of the deposited magnetic layers was adjusted by controlling the number of the Zr chips. During sputtering, an Ar flow rate of 20 SCCM (SCCM denotes cubic centimeters per minute at STP) was needed to maintain an Ar pressure of 0.15 Pa, and the rf power density was 1.7 W/cm^2. The structures of $(Co_{96}Zr_4/Cu)_n$ multilayer thin films with a Cu buffer layer was shown in Fig. 1. Thickness of total $Co_{96}Zr_4$ layers is 180 nm and each Cu layer is 20 nm, which was used to eliminate the exchange interaction between $Co_{96}Zr_4$ layers.

The three following parameters need to be determined: saturation magnetization M_s, static permeability μ_s and resonance frequency f_r. M_s were determined by $m_s/(St)$, where m_s is the saturation magnetic moment measured by vibrating sample magnetometer (Lakeshore model 7304), and S and t are the area and thickness of magnetic layers, respectively. $\mu_s - 1$ were determined by $\mu_s - 1 = 4\pi M_s/H_k$, where H_k was the in-plane uniaxial magnetic anisotropies determined by calculating the measured easy axis and hard axis loops of the reduced magnetization.[9] f_r were determined by fitting the permeability spectrum, which were carried out with a PNA E8363B vector network analyzer using the microstrip

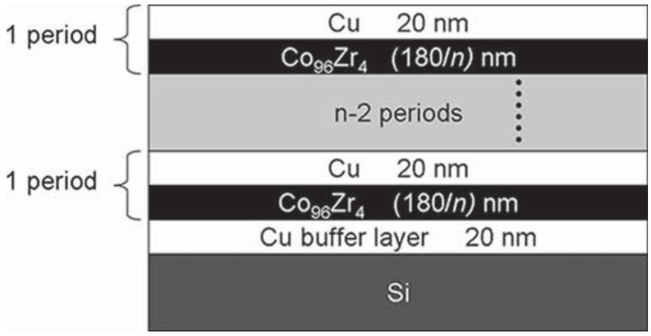

FIG. 1. The structure of $(Co_{96}Zr_4/Cu)_n$ multilayers on a 20 nm Cu buffer layer. The substrates are $10\times20\times0.42$ mm^3 (111)-oriented Si pieces. Thickness of each $Co_{96}Zr_4$ layers are $(180/n)$ nm. Between $Co_{96}Zr_4$ layers, there are 20 nm Cu layers to eliminate the exchange interaction.

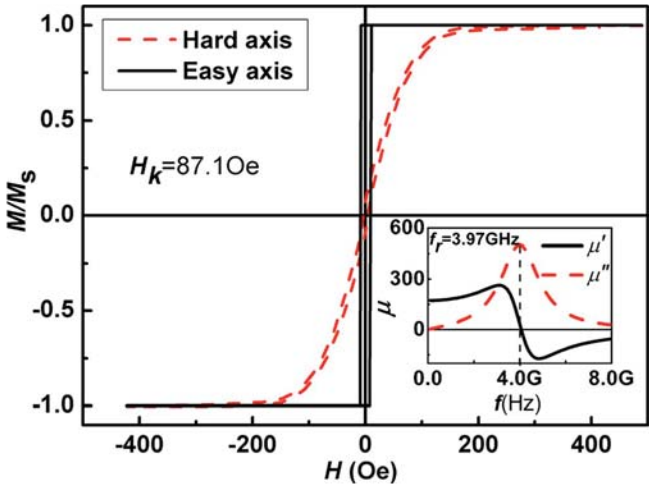

FIG. 2. (Color online) The main figure is the in-plane M-H loop, where red dashed line is the M-H loop of hard axis and the blank solid line is the M-H loop of easy axis. The inset is the permeability spectrum of $(Co_{96}Zr_4/Cu)_{24}$ multilayers, where red dashed line and the solid line show the image part and the real part of the permeability.

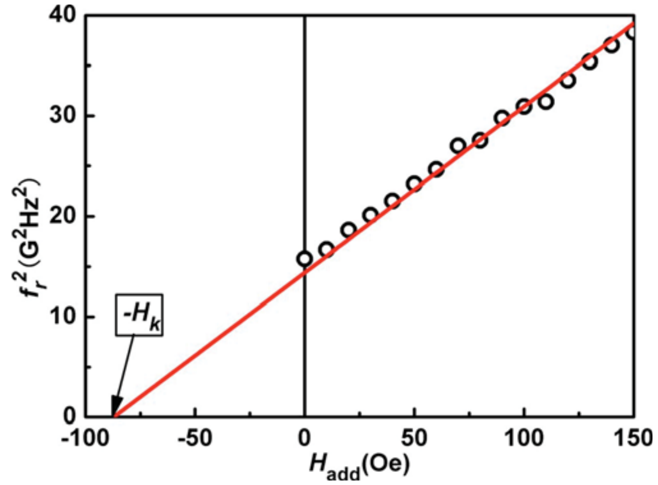

FIG. 3. (Color online) Square of the resonance frequencies for additional field H_{add} parallel to the easy axis are shown. The circles are experimental results and the red line is the fitting curve. The sample is $(Co_{96}Zr_4/Cu)_{24}$ multilayers. The value of H_{add} at the zero crossing of f_r^2 is 87.0 Oe.

method from 100 MHz to 8 GHz with sample ($5\times5\times0.42$ mm^3) positioned in the middle of strip line with inner height 0.8 mm between upper line and ground plate, width of upper line 3.94 mm, length 9 mm.[12] One of the static and dynamic results of $(Co_{96}Zr_4/Cu)_n$ multilayers are shown in Fig. 2. These samples were well in-plane uniaxial anisotropic thin soft magnetic film from the static magnetic hysteresis loop.

In order to prove that the results of f_r are natural mode resonance frequencies, we measured the different resonance frequencies f_r for an additional field H_{add} parallel to the easy axis of the films. For natural resonance, with an additional field H_{add}, the square of f_r should have a linear relationship with the additional field, and the linear extrapolation of f_r^2 should cross the H_{add} axis with the same value as $-H_k$.[7] In Fig. 3, the square of f_r as a function of H_{add} are shown. The good agreement between the fitting line and the results of experiment proved that the resonance results of $(Co_{96}Zr_4/Cu)_{24}$ multilayers by using the microstrip method are exactly the natural resonance.

The results of anisotropy field H_k, the static magnetic susceptibility μ_s-1, measured resonance frequency f_r, the calculated results of resonance frequency f_c by the Kittel equation, and Acher's limits of $(Co_{96}Zr_4/Cu)_n$ are shown in Table I, where $4\pi M_s=15$ kg of $Co_{96}Zr_4$ thin films are used. The results showed that the experimental resonance frequency f_r are much larger than calculated results of resonance frequency f_c for the samples which has large number of periods. Moreover, it is the same similar behavior for Acher's limit (e.g., $n=30$ or $n=40$ as facts of 1.66, 1.76, etc.).

It is known that the effects of dipole interaction and exchange couple and in-plane stress should be eliminated because these interactions can be shown in static magnetic hysteresis loops. The significant difference between single layer and multilayers is that the number of interfaces has been enlarged. So, the possible reason of the extended Acher's limit in $(Co_{96}Zr_4/Cu)_n$ multilayers may be the interface interactions. If interface interactions were considered in the dynamic magnetic behavior, the total free energy density of each magnetic layer should be explicitly written as

$$E = K_1 \sin^2 \varphi + \frac{1}{2} N_z M_s^2 \cos^2 \theta + \frac{2K_u}{t} \sin^2 \theta, \quad (4)$$

where K_1 and K_u are the uniaxial in-plane anisotropy constant and interface anisotropy constant, $N_z=4\pi$ is the demagnetizing coefficient of the perpendicular direction of thin films, t is the thickness of magnetic single layer, θ is the angle between perpendicular direction and the magnetic moment, and φ is the angle between easy axis and the magnetic moment. The energy density of interface used here was within Néel's model.[13] The interface anisotropy is out of plane anisotropy for positive interface anisotropy constant or in-plane anisotropy for negative interface anisotropy con-

TABLE I. The value of anisotropy field H_k, resonance frequency f_r which were analyzed from experimental result, the results of resonance frequency f_c which were calculated from H_k using Kittel equation, μ_s-1 and Acher's limits of $(Co_{96}Zr_4/Cu)_n$ are shown.

Sample with different periods ($4\pi M_s=15$ kg, $L_A=1764$ G^{2}Hz^2)	H_k (Oe)	f_c (GHz)	f_r (GHz)	μ_s-1	$(\mu_s-1)\,f_r^2$ (G^2 Hz^2)
$(Co_{96}Zr_4/Cu)_1$	32.0	1.96	1.98	469	1838
$(Co_{96}Zr_4/Cu)_{12}$	66.3	2.50	3.12	226	2202
$(Co_{96}Zr_4/Cu)_{15}$	142.8	3.66	4.59	105	2213
$(Co_{96}Zr_4/Cu)_{18}$	133.3	3.54	4.55	112	2330
$(Co_{96}Zr_4/Cu)_{24}$	87.1	2.86	3.97	172	2687
$(Co_{96}Zr_4/Cu)_{30}$	116.6	3.31	4.77	129	2927
$(Co_{96}Zr_4/Cu)_{40}$	114.4	3.28	4.86	131	3096

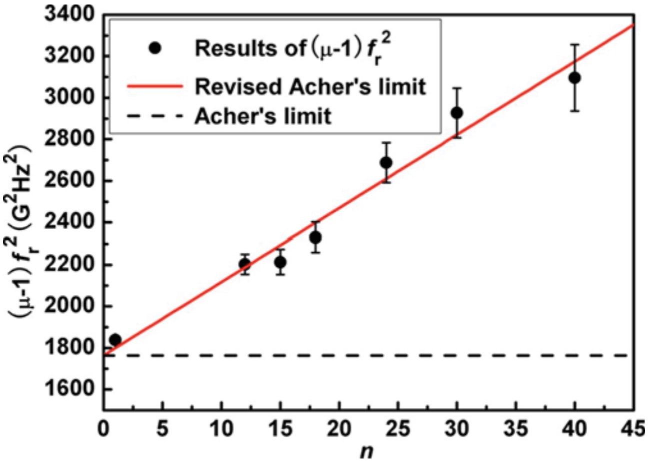

FIG. 4. (Color online) The relationship between Acher's limit and number of periods is displayed. The solid cycles are results calculated from experimental resonance frequency with function $(\mu_s-1)\ f_r^2$, where the red solid line is fitting curve of RAL and the blank dashed line is the Acher's limit of thin film.

stant. With the Landau–Lifshitz equation[14] and Eq. (4) the resonance frequency is

$$f_r' = \frac{\gamma}{2\pi}\sqrt{H_K\left(4\pi M_s - 2\frac{2K_u}{M_s t}\right)}, \tag{5}$$

and the Acher's limit should be rewritten as revised Acher's limit (RAL)

$$L_A' = (\mu_s - 1)f_r^2 = \left(\frac{\gamma}{2\pi}\right)^2 4\pi M_s\left(4\pi M_s - 2\frac{2K_u}{M_s t}\right). \tag{6}$$

The results of experiment (marked with solid circles), Acher's limit (marked with blank dashed line), and RAL (marked with red solid line) are shown in Fig. 4. This figure shows that the RAL has a linear relationship with the number of periods n instead of being constant, which results from the negative value of derived K_u as $-1.61\ \mathrm{erg/cm^2}$. The negative interface anisotropy constant K_u of Co/Cu $(11n)$ interface was also found by Chuang *et al.*[15] So the interface anisotropy of $Co_{96}Zr_4$/Cu interface is in-plane isotropic anisotropy. The well fitting of RAL and experimental data reveals that considering the effect of interface interaction is necessary to study the relationship of permeability and resonance frequency of multilayers.

Figure 5 shows analyzed, previously published datum for high frequency single layer soft magnetic films such as CoNb,[10] CoAlO,[16] and $(Co_{96}Zr_4/Cu)_n$ multilayers with three lines showing the Acher's limit of CoNb thin film (marked with red dot line) and the RAL of $Co_{96}Zr_4$ single layer with Cu buffer and cover layer (marked with green dashed line) and $(Co_{96}Zr_4/Cu)_n$ multilayer for $n=40$ (marked with blank solid line). In this figure, it can be seen that the resonance frequencies of traditional single thin film are restricted by the Acher's limit. However, for $(Co_{96}Zr_4/Cu)_n$ thin films, resonance frequencies obey the RAL instead of the Acher's law. Moreover, the thinner the thickness of each layer, the larger the RAL of $(Co_{96}Zr_4/Cu)_n$ multilayers. So, the RAL of $(Co_{96}Zr_4/Cu)_n$ multilayers could exceed the Acher's limit of all soft magnetic thin films including $Fe_{65}Co_{35}$ thin film [largest Acher's limit of thin film with $4\pi M_s=24.5$ kg (Ref. 17)] if thickness of each layer is small enough.

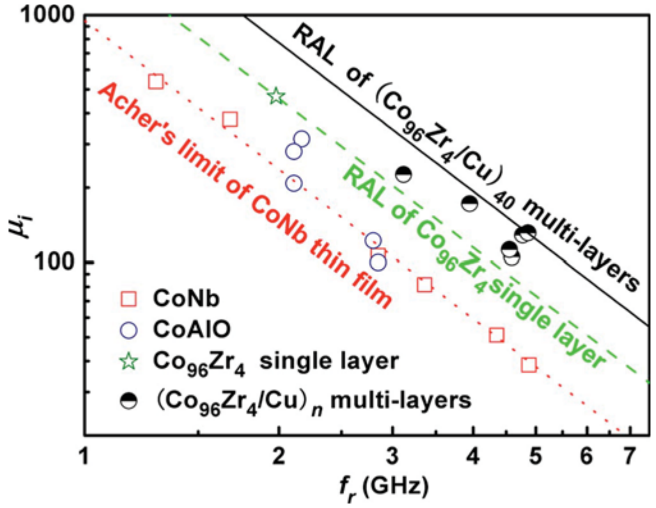

FIG. 5. (Color online) Initial permeabilities as a function of the resonance frequency from room temperature, high frequency studies of Co based soft magnetic films in recent published papers are shown. The half solid circles are results from the present work. The blank solid line, the green dashed line and the red dot line represent the RAL of $(Co_{96}Zr_4/Cu)_{40}$ multilayer with negative value K_u, RAL of $Co_{96}Zr_4$ single layer and Acher's limit of CoNb thin film.

In summary, the $(Co_{96}Zr_4/Cu)_n$ multilayers have good high frequency properties. The larger Acher's limit than single layer films were resulted by the negative interface anisotropy of $Co_{96}Zr_4$/Cu. With increasing number of interface, for same μ_s and H_k, f_r of the multilayers increases, which can enlarge the working frequency range. This work is very useful for searching new materials with high permeability at high frequency.

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