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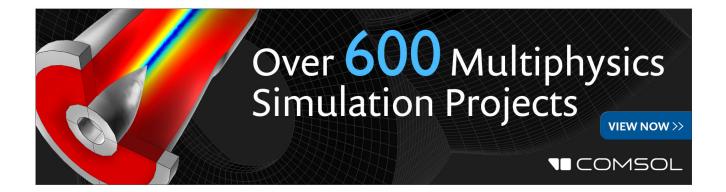
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Tunable asymmetric magnetoimpedance effect in ferromagnetic NiFe/Cu/Co films

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We investigate the magnetization dynamics through the magnetoimpedance effect in ferromagnetic NiFe/Cu/Co films. We observe that the magnetoimpedance response is dependent on the thickness of the non-magnetic Cu spacer material. We verify asymmetric magnetoimpedance in films with biphase magnetic behavior and explore the possibility of tuning the linear region of the magnetoimpedance curves around zero magnetic field by varying the thickness of the spacer and probe current frequency. We discuss the experimental results in terms of the different mechanisms governing the magnetization dynamics at distinct frequency ranges, quasi-static magnetic properties, thickness of the spacer, and the kind of the magnetic interaction between the ferromagnetic layers. The results place films with biphase magnetic behavior exhibiting asymmetric magnetoimpedance effect as very attractive candidates for application as probe element in the development of auto-biased linear magnetic field sensors. © 2014 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4895708]

The magnetoimpedance effect (MI), known as the change of the electrical impedance of a ferromagnetic conductor submitted to an external static magnetic field, is commonly employed as a tool to investigate magnetic materials. In recent years, the interest on this effect has grown due to its contribution to the understanding of physics associated to magnetization dynamics² and to the possibility of application of magnetic materials as probe element in sensor devices for low-field detection.³ Experiments have been carried out in numerous magnetic systems, including ribbons, 4,5 wires, 6-9 and films. 10-18 However, although soft magnetic materials are highly sensitive to small field variations at low magnetic fields, most of them have essentially nonlinear MI behavior around zero magnetic field, which prevents a simple straightforward derivation of an appropriate signal for sensor applications. 18,19

The shift of the sensor operational region and the leading of the linear MI behavior at around zero field are primarily obtained by applying a bias field or an electrical current to the ordinary MI element. However, this approach proved to be disadvantageous from practical point of view, mainly due to energetic consumption. Recently, it has been shown that materials exhibiting asymmetric MI (AMI) effect arise as promising alternative for applications, opening possibilities for the use of this kind of materials in auto-biased linear magnetic field sensors. For these materials, the asymmetric effects are obtained by inducing an asymmetric static magnetic configuration, usually done by magnetostatic interactions 9,19,21–23 or exchange bias, 11,18,24,25 or by playing with the orientation between the external magnetic field and anisotropy. 26

For films, the primary AMI results have been measured for exchange biased multilayers. Theory and experiment showed MI curves shifted by the exchange bias field

and verified that the linear region of AMI curves can be

In this work, we investigate the magnetoimpedance effect in ferromagnetic NiFe/Cu/Co films. We observe that the MI response is dependent on the thickness of the Cu spacer material, a fact associated to the kind of the magnetic interaction between the ferromagnetic layers. We show that the linear region of the AMI curves in films with biphase magnetic behavior is experimentally tunable by varying the thickness of the spacer and probe current frequency. The results place films with biphase magnetic behavior exhibiting AMI effect as very attractive candidates for application as probe element in the development of auto-biased linear magnetic field sensors.

For this study, we produce $Ni_{81}Fe_{19}(25 \text{ nm})/Cu(t_{Cu})/$ Co(50 nm) films, with $t_{Cu} = 0$, 1.5, 3, 5, 7, and 10 nm. The films are deposited by magnetron sputtering onto glass substrates, with dimensions of $8 \times 4 \,\mathrm{mm}^2$, using the following parameters: base vacuum of 10⁻⁸ Torr, deposition pressure of 2.0 mTorr with Ar at 32 sccm constant flow, and 150 W (DC) for NiFe and Co layers, while 100 W (RF) for Cu layer. During the deposition, a constant 2 kOe magnetic field is applied perpendicularly to the main axis of the substrate to induce a magnetic anisotropy and define an easy magnetization axis. X-ray diffraction results indicate the Co(111) and NiFe(111) preferential growth of all films. Magnetization curves are measured with a VSM along and perpendicular to the main axis of the films. Magnetization dynamics are investigated through MI measurements obtained using a RFimpedance analyzer Agilent model E4991, with E4991A test

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tuned to around zero just by modifying the angle between applied magnetic field and exchange bias field, or changing the probe current frequency. However, another promising possibility of AMI material resides in films presenting biphase magnetic behavior, with hard and soft ferromagnetic phases intermediated by a non-magnetic layer acting together.

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FIG. 1. Representative normalized magnetization curves for selected NiFe/Cu/Co films with different t_{Cu} , measured with external in-plane field applied along (||) and perpendicular (\perp) to the main axis. Films with $t_{Cu} < 3$ nm present behavior similar to that for the film without spacer, while the films with $t_{Cu} > 3$ nm have behavior identical to that observed for the film with $t_{Cu} = 7$ nm. The film with $t_{Cu} = 3$ nm is within the critical Cu thickness range and presents an intermediate behavior.

head connected to a microstrip, following the procedures traditionally employed by our group. ^{13,16,17} To quantify the sensitivity as a function of the frequency, we calculate the magnitude of the impedance change at low fields through ¹⁸

$$\frac{|\Delta Z|}{|\Delta H|} = \frac{|Z(H = 6 \text{ Oe}) - Z(H = -6 \text{ Oe})|}{12}.$$
 (1)

We consider $|\Delta Z|$ since the impedance around zero field can present positive or negative slopes, depending on the sample and frequency. It is verified that $|\Delta Z|/|\Delta H|$ is roughly constant at least for a reasonable low field range.

Figure 1 shows the magnetization curves for selected films. When analyzed as a function of t_{Cu} , an evolution of the shape of the magnetization curves is observed, indicating the existence of a critical thickness range, ~ 3 nm, which splits the films in groups according the magnetic behavior. For films with $t_{Cu} < 3$ nm, the NiFe and Co layers are ferromagnetically coupled. The angular dependence of the curves indicates an uniaxial in-plane magnetic anisotropy, induced by the field applied during the deposition. Despite the similar magnetic behavior, the film with $t_{Cu} = 1.5 \text{ nm}$ has slightly higher coercive field if compared to that for the film without spacer, possibly associated to the increase of the whole sample disorder due to the non-formation of a regular complete Cu layer. ¹³ The film with $t_{Cu} = 3$ nm presents an intermediate magnetic behavior, with smaller magnetic permeability, characterized by the first evidences of a small plateau, and the appearance of magnetization regions associated to distinct anisotropy constants of the NiFe and Co layers. Films with $t_{Cu} > 3$ nm exhibit a biphase magnetic behavior. The two-stage magnetization process is characterized by the magnetization reversion of the soft NiFe layer at low field, followed by the reversion of the hard Co layer at higher field. None substantial difference between the curves is verified. In principle, the biphase magnetic behavior suggests that the ferromagnetic layers are uncoupled. The easy magnetization axis remains perpendicular to the main axis of the substrate, as expected. The weaker anisotropy induction and increase of hysteretic losses are primarily related to the roughness of the interfaces and lack of homogeneity of the Cu layer arisen as its thickness is raised. 13

It is well-known that quasi-static magnetic properties play a fundamental role in the dynamic magnetic response and MI behavior. ¹⁸ The shape and amplitude of the MI curves are strongly dependent on the orientation of the applied field and *ac* current with respect to the magnetic anisotropies, magnitude of the external field, and probe current frequency as

well as are directly related to the mechanisms responsible for the transverse magnetic permeability changes: skin and ferromagnetic resonance (FMR) effects. ^{18,27,28} However, MI effect can also provide further insights on the nature of the interactions governing the magnetization dynamics and energy terms affecting the transverse permeability.

Regarding the MI results, Fig. 2 shows the curves, at a selected frequency, for films with different t_{Cu} . All samples exhibit a double peak behavior for the whole frequency range, a signature of the perpendicular alignment of the external field and ac current with the easy magnetization axis. An interesting feature related to the MI behavior resides in the dependence of the amplitude and position of the peaks with t_{Cu} , and the probe current frequency.

Films with t_{Cu} < 3 nm present the well-known symmetric MI behavior for anisotropic systems. The curves have the double peak behavior, symmetrical at around H = 0, the peaks having roughly the same amplitude. For frequencies up to \sim 0.85 GHz, the peaks position remains unchanged

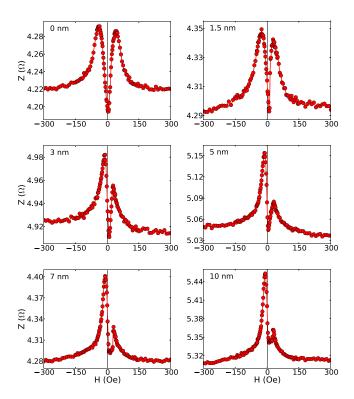


FIG. 2. The MI curves at frequency of 0.75 GHz measured for the films with different t_{Cu} . The curves are acquired over a complete magnetization loop and present hysteretic behavior. We show just the curve with increasing field.

close to the anisotropy field, indicating the skin effect as the main responsible by the changes of transverse permeability. For frequencies above this value, not presented here, besides the skin effect, the FMR effect also becomes an important mechanism responsible for the MI variations, a fact evidenced by the displacement of the peaks position toward higher fields as the frequency increases. The contribution of the FMR effect to Z is also confirmed using the method described by Barandiarán *et al.*²⁹ and previously employed by our group. ¹²

However, films with $t_{Cu} \ge 3$ nm present noticeable asymmetric magnetoimpedance effect. The asymmetric behavior is assigned by two characteristic features: shift of the MI curve in field, depicted by the asymmetric position of the peaks, and asymmetry in shape, evidenced by the difference of amplitude of the peaks. Figure 3 shows the evolution of the MI curves, at selected frequencies, for the film with $t_{Cu} = 7$ nm, as an example of the results obtained for films with biphase magnetic behavior. Regarding the position of the peaks, since the skin effect commands the dynamical behavior, the peaks position remains invariable at low and intermediate frequencies. For this film, the peak at negative field is located at ~ -4 Oe, while the one at positive field is at $\sim +30$ Oe. For the other films with $t_{Cu} > 3$ nm, the peak at positive field is placed at similar value, although the location of that at negative field presents dependence with t_{Cu} , as will be discussed. With respect to the amplitude of the peaks at low and intermediate frequencies, for all films with $t_{Cu} > 3$ nm, the peak at negative field has higher amplitude than that at positive field. As a signature of the emergence of the FMR effect, the displacement of the peak at negative field begins at ~0.6 GHz, while the position of peak at positive field starts changing at ~ 1.1 GHz. Above ~ 1.5 GHz,

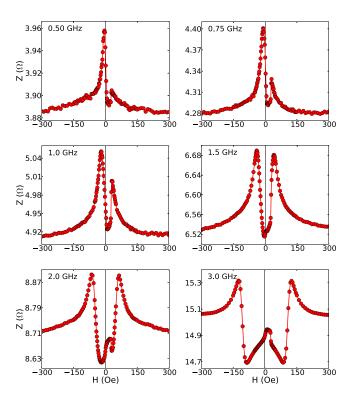


FIG. 3. The MI curves for selected frequencies for the film with $t_{Cu} = 7$ nm. Similar results are obtained for all films with biphase magnetic behavior.

strong skin and FMR effects are responsible by the MI variations. At this high frequency range, the asymmetry still remains in the portion of the impedance curve around the anisotropy fields. However, the displacement of the peaks toward higher fields suppresses the peak asymmetry, in position and amplitude, resulting in symmetric peaks around H=0 with same amplitude. For the film with $t_{Cu}=3$ nm, similar features are observed, respectively, at ~ 0.75 GHz, ~ 1.1 GHz, and ~ 2.0 GHz.

The most striking finding resides in the asymmetry in the MI curves measured for films with biphase magnetic behavior. It is important to notice that the MI response is nearly linear for the range of low fields, and the shape of the Z curves depends on the t_{Cu} and probe current frequency. As a consequence, the best response can be tuned by playing with both parameters. Figure 4 shows the frequency spectrum of impedance variations, defined by Eq. (1), for each film, indicating the sensitivity around zero field. From the figure, we verify that the films split in different groups according to the sensitivity, each one related to a given magnetic behavior, verified through the magnetization curves. Films with $t_{Cu} < 3$ nm have the largest sensitivity values at \sim 1.0 GHz, the film with t_{Cu} = 3 nm at \sim 0.9 GHz, while the ones with $t_{Cu} > 3$ nm at ~ 0.75 GHz. For all of them, the sensitivity peak is found to be at frequencies just after the FMR effect starts appearing. The highest sensitivity is observed for the films with $t_{Cu} > 3 \text{ nm}$, reaching $\sim 8 \text{ m}\Omega/\text{Oe}$, and seems to be insensitive to t_{Cu} .

Our results raise an interesting issue on the MI behavior and the energy terms affecting the transverse magnetic permeability. Generally, our films consist of two ferromagnetic layers, with distinct anisotropy fields, intermediated by a non-magnetic spacer material. We interpret our experimental data as a result of the competition between two kinds of magnetic interactions between ferromagnetic layers: ferromagnetic exchange coupling and magnetostatic coupling. They are strongly dependent on the spacer thickness and affect in different ways the MI behavior.

For t_{Cu} < 3 nm, the strong coupling is due to the exchange interaction between touching ferromagnetic layers or through pinholes in the non-magnetic spacer. The

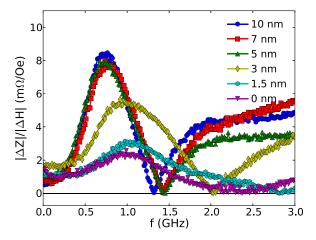


FIG. 4. Frequency spectrum of impedance variations between ± 6 Oe for the films with different t_{Cu} , indicating the sensitivity around zero field. Notice that the saturation effect observed as t_{Cu} increases above 3 nm, related to the amplitude and frequency at which the maximum sensitivity is reached.

coupling adjusts parallelly the magnetizations of the NiFe and Co layers, and the whole sample behaves as a single ferromagnet. 13 As expected, we confirm for them the symmetric MI behavior of single anisotropic systems. For $t_{Cu} > 3$ nm, the Cu layer is completely filled, 13 and the nature of the coupling is magnetostatic. If the ferromagnetic layers were completely uncoupled, one could expect multiple peaks MI behavior, associated to the anisotropy fields of the layers. This is not verified here, indicating that the AMI cannot be explained assuming independent reversal of the NiFe and Co layers. Thus, the asymmetry arises as a result of the magnetostatic coupling between the ferromagnetic layers. The origin of this coupling is ascribed to the hard Co magnetic phase in terms of an effective bias field, induced by divergences of magnetization mainly due to roughness in the interfaces and limits of the sample 19 that must be taken into account as a contributor to the transverse permeability. The field penetrates the non-magnetic spacer and acts on the magnetization of the soft NiFe layer. It is important to point out that the anisotropy field of the Co layer is considerable larger than that of the NiFe layer, the reason why this asymmetric behavior is not verified in traditional multilayers.

In this sense, the main features of the AMI verified in films with biphase magnetic behavior can be explained through the effective interaction between the ferromagnetic layers. The influence on the NiFe layer is dependent on the magnetic state of the Co layer, as well as on the t_{Cu} . The difference of amplitude of the peaks is understood in terms of the orientation of the magnetization of the two layers. The peak at negative field is higher than that at positive field, since the magnetization of the NiFe layer is parallel to the one of the Co layer, and to the magnetostatic and external fields. Once the magnetization of the NiFe layer is reverted as the field increases, its sense with respect to the magnetostatic field is modified, and this form closes the magnetic flux, resulting a lower peak. 19 AMI with similar features has also been verified in field-annealed Co-based amorphous ribbons. 22,23 When the MI curve for decreasing magnetic field is considered, the reverted behavior is verified, due to the opposite sense of the magnetization of the Co layer.

By employing MI effect, the effective coupling strength between the NiFe and Co layers can be estimated. This can be done by considering the location of the MI peaks at low and intermediate frequencies. Figure 5 shows the magnetic field values in which the impedance peaks are located, for different t_{Cu} . The position of the peak at negative field has a noticeable dependence with t_{Cu} . In particular, it is verified a reduction of the field value where the peak is located as t_{Cu} increases, corroborating the assumption of a magnetostatic origin of the coupling between the ferromagnetic layers. We interpret this reduction as an indication of the decrease of the bias field intensity acting on the NiFe layer as the Cu spacer becomes thicker. On the other hand, the peak at positive field is located at \sim 30 Oe, except for the sample without spacer. The constancy, irrespective of t_{Cu} and field value of the Co reversion for each sample, suggests that this value corresponds to an intrinsic feature of the ferromagnetic Co layer, since it has similar thickness for all samples. Thereby, we understand it as the magnitude of the bias field induced by the Co layer.

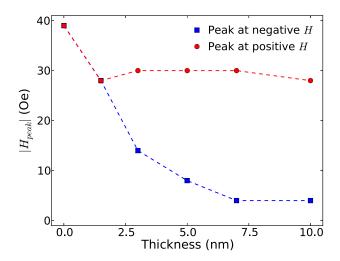


FIG. 5. Magnetic field values in which the impedance peaks are located, at 0.5 GHz, for the films with different t_{Cu} . The values are obtained from MI curves acquired when the field goes from negative to positive values. Notice that films with $t_{Cu} \ge 3$ nm present AMI behavior.

In conclusion, we have investigated the MI effect in ferromagnetic NiFe/Cu/Co films and observed the dependence of the curves, amplitude, and position of the peaks, with t_{Cu} . We have verified that the MI response of these films can be tailored by the kind of magnetic interaction between the ferromagnetic layers. From the results, we have observed the crossover between two distinct magnetic behavior, governed by exchange interaction or magnetostatic coupling, as the Cu thickness is altered crossing through $t_{Cu} = 3$ nm. We have tuned the linear region of the AMI curves around zero magnetic field by varying t_{Cu} and probe current frequency. The highest sensitivity is observed for films with $t_{Cu} > 3 \text{ nm}$, reaching ~ 8 m Ω /Oe, and seems to be insensitive to t_{Cu} . It is known that the employment of multilayers enables us to reach higher sensitivity values. In this sense, experiments in multilayered ferromagnetic biphase films are currently in progress. Thus, these results extend the possibilities for application of ferromagnetic films with AMI as probe element for the development of auto-biased linear magnetic field sensors, placing films with biphase magnetic behavior as promising candidates to optimize the MI sensitivity.

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