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BP-09

Anisotropy and damping of molecules/cobalt hybrid thin films

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We have investigated the magnetic properties of evaporated Co thin films covalently functionalized with different organic thin films, namely (1-(2-bisthienyl benzene) and nitro-benzene. The coating is realized thanks to a diazonium-based electro-reduction process. Brillouin light scattering experiments revealed that the magnetic properties are sensitive to the presence of the organic film. For (1-(2-bisthienyl benzene) thin films, the perpendicular magnetic anisotropy is increased as well as the magnetic damping. However for nitrobenzene, only the perpendicular anisotropy is increased albeit less than for (1-(2-bisthienyl benzene). This change in magnetic properties might be attributed to the coupling with the organic molecules.

***Index Terms*— Anisotropy, Damping, Organic molecules, Cobalt thin films, Brillouin light scattering.**

I. INTRODUCTION

In the last decades, there is a considerable effort in research to control the properties of magnetic materials and spins of electrons for spintronics applications as memories [1-7]. The classical method is to combine magnetic thin layers with different properties or to tune an alloy composition [3, 4, 8, 9]. Another emerging possibility is the coupling of a ferromagnet with organic molecules. Organic molecules can affect the surface magnetic properties mainly through interfacial effects [2, 5, 6, 10-13]. Currently, the achievable thickness of the ferromagnetic layers is thin enough, in nm range, to achieve enhancement of the magnetic properties through the interfacial interaction. The interest of organic layers is the variety of means to reliably add them in devices. The most used deposition techniques are the evaporation [1], the self-assembly of molecular monolayers [2] and the electrochemical grafting (electrochemical coating) [14]. In case of electrochemical coating, the organic molecule is covalently bonded to the ferromagnetic layer's surface. Stronger coupling [15] and thus tuning of the properties is expected compared to the physisorption case obtained by evaporation. *Ab-initio* calculations [10, 16, 17] demonstrate that what is important is the delocalized electrons as the conduction band of graphene [18]. In molecules, this corresponds to the π-bands which tend to strongly hybridize at the surface with a metal. Adsorption of molecules on a ferromagnetic surface can also induce molecular roughening causing substantial changes in anisotropies and spin polarization [19]. However, the experimental characterization of magnetization dynamics when coupled to organic molecules has to be done for development of organic spintronic devices. A well-known non-invasive tool for probing magnetization dynamics is the Brillouin light scattering (BLS) [20-22].

In this paper, we report measurements of BLS spectra of a Co layer coated by either 1–(2-bisthienyl) benzene (BTB) or nitro benzene (NB) through the same diazonium-based electrochemistry process. We found that the magnetic properties are strongly tuned when the molecule is rich in π-electrons. Our results demonstrate that the magnetic anisotropy as well as the damping can be tuned by the appropriated organic molecule.

II. EXPERIMENTAL

Chemicals: 1-(2-bisthienyl)-4-aminobenzene (BTB) was synthesized according to published procedures [23]. All chemicals were purchased from Sigma Aldrich and used as received. Lithium perchlorate ($LiClO_4$) and tetrabutylammonium tetrafluoroborate (Bu_4NBF_4) were used as supporting electrolyte at 0.1 M concentration in acetonitrile (ACN).

Ti (1 nm)/Pt (20 nm)/Co (8 nm) heterostructures were deposited by electron-beam evaporation under high vacuum conditions (10^{-8} mBar) on Si/SiO_2 substrate. Low deposition rates (0.05 nm/s) were chosen.

For the electrochemical experiments a conventional three-electrode cell was used. Platinum wire was used as auxiliary electrode, saturated calomel electrode (SCE) was used as reference electrode. Before any electrochemical measurements the solutions were deoxygenated by bubbling argon gas for 30 minutes, during the experiment the electrochemical cell remain under argon. The potentiostat used in this study was a CHI 660C (CH Instruments, made in USA).

Electrochemical deposition of the organic layer: Before electrochemical coating, the cobalt electrodes were immersed in an acidic solution and a negative potential was applied

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during 10 seconds to remove the CoO oxide layer. The cobalt layer was then modified by either 1–(2-bisthienyl) benzene (BTB) or nitro benzene (NB) through the same diazonium-based electrochemistry process. In the case of NB, a solution of 0.5 mM of nitrobenzene diazonium salt (NBD) in ACN with 0.1 M Bu_4NBF_4 was prepared for the electrochemical coating [24, 25]. In the case of BTB, on the contrary, the formation of the diazonium cation from BTB precursor was performed *in situ* [26]. Briefly, an ACN solution containing of 1-(2-bisthienyl)-4-aminobenzene precursor (0.5 mM) and Bu_4NBF_4 (0.1 M) as supporting electrolyte was prepared and degassed by argon bubbling for at least 10 minutes. Following that, tBuO-NO (30 equivalents) was added in excess to the solution. As the reaction between amine function and tert-butyl nitrite is relatively slow, coating was only started after 5 minutes incubation time. NB and BTB layer deposition was performed on glassy carbon electrodes as well as on cobalt electrodes using cycling voltammetry (Fig. 1). The number of cycles gives us a thickness of around 7 ± 0.6 nm for both depositions.

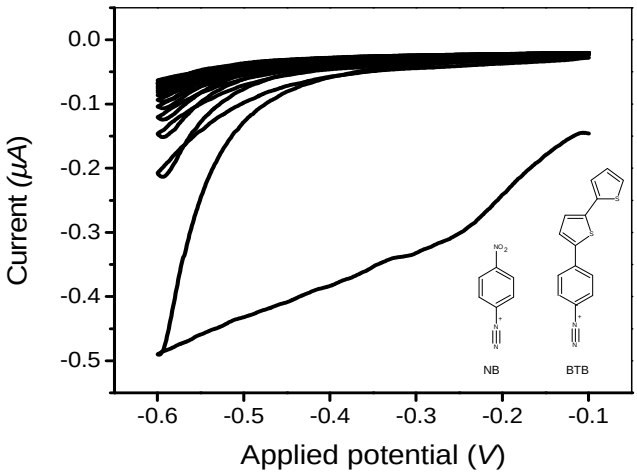

Fig.1: Cyclic voltammetry on cobalt substrate for the reduction of NB diazonium salt (5×10^{-4} mol L^{-1}) in ACN with 0.1 M of supporting electrolyte. 10 cycles at scan rate 0.1 $V.s^{-1}$, inset: structures of NB and BTB diazonium salts.

After the coating of the molecules, we measured the magnetization dynamics behavior of the samples using a conventional BLS technique.

The BLS technique gives access to magnon modes with nonzero wavevector values. In the BLS experiment, the magnons of a wavenumber k in the range 0–20 μm^{-1} (depending on the incidence angle θ: in backscattering configuration), are probed by illuminating the sample with a laser having a wavelength λ=532 nm. The laser power has been adapted in order to avoid any irremediable degradation of the molecules. The magnetic field was applied perpendicular to the incidence plane, which allows for probing excitons propagating along the in-plane direction perpendicular to the applied field: Damon-Eshbach (DE) geometry. The Stokes (S, negative frequency shift relative to the incident light as a magnon was created) and anti-Stokes (AS, positive frequency shift relative to the incident light as a magnon was absorbed) frequencies, are detected simultaneously. BLS spectra of ferromagnetic films can be characterized by a pronounced Stokes/Anti-Stokes intensity asymmetry that is known to be a peculiarity of time-reversal symmetry and that is reversed under the reversed bias. The probed magnons were studied as a function of applied magnetic field (up to 200 mT). Frequencies and the peaks linewidth (full width at half maximum: *FWHM*) were determined from Lorentzian fits to the BLS spectra.

III. Results and Discussions

A. Nitrobenzene (NB) measurements

We measured BLS spectra of the non-coated and NB coated areas of the Co film, with an incident angle of 35°, corresponding to a wavenumber k of 13.5 μm^{-1}. Figure 2 shows typical obtained raw spectra (Stokes side). The magnons peaks were visible in both areas of the sample. In the Co area, the magnons peak is at higher frequency than in the coated area. It is to mention that we hardly observe magnons peak in the large area where the coating is fully achieved. This point is under investigations.

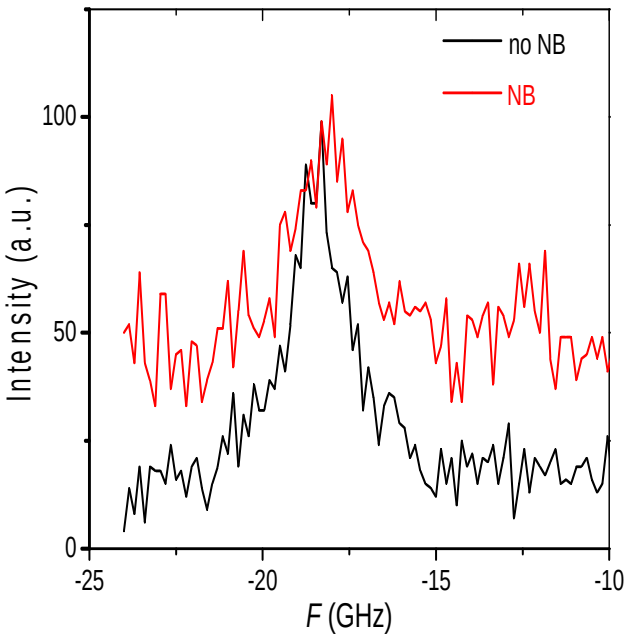

Fig. 2 (Colour online) Raw BLS spectra (Stokes side) obtained under a 200 mT in plane magnetic field and a wavenumber k equal to 13.5 μm^{-1}; black: Co area (with presence of CoO); red: area coated with NB molecules.

Figure 3 exhibits the experimental magnons frequency (fig. 3a) and the peaks linewidth *FWHM* (fig. 3b) evolution as function of the external magnetic field. As expected, the measured frequencies on the two areas are clearly shifted for all used values of magnetic field.

The magnons frequency is given by the dispersion law [27]:

$$F = \mu_0 \frac{\gamma}{2\pi}\sqrt{\left(H + Jk^2 + P(kt)M_s\right)\left(H + Jk^2 - P(kt)M_s + M_s - H_K\right)} \quad (1)$$

where H is the in-plane applied field, M_s is the saturation magnetization, t is the ferromagnetic layer thickness, γ is the gyromagnetic coefficient $\gamma/(2\pi)$ = 30 GHz/T, μ_0 is the permeability of vacuum, $J = 2A_{ex}/\mu_0 M_s$, with A_{ex} the exchange stiffness constant of the magnetic material, H_K is the

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perpendicular uniaxial anisotropy field ($H_K = 2K/M_s$, where K is the anisotropy constant), and $P(kt) = 1 - \frac{1-\exp(-kt)}{kt}$. The coefficient $P(kt)$, describing dipolar interactions, reduces in thin films ($kt << 1$) to a simple $kt/2$, which makes this term linear in kt. It should be noticed that $kt \approx 0.11$ in our case. It is to notice that the frequency of the probed Damon-Eshbach (DE) surface mode given by (1) is largely insensitive to whether the exchange constant A_{ex} is assumed or fitted.
A change of frequency, at fixed k wavenumber and magnetic field H, should correspond to either a change in the magnetic anisotropy or a change in the Co thickness. The question is to know which one is dominant as potential factor in this change. This is discussed later in this paper. Concerning the magnons damping, the peaks linewidth *FWHM*, represented in fig. 3b, did not show change, within the considered experimental uncertainties, between the two investigated areas.

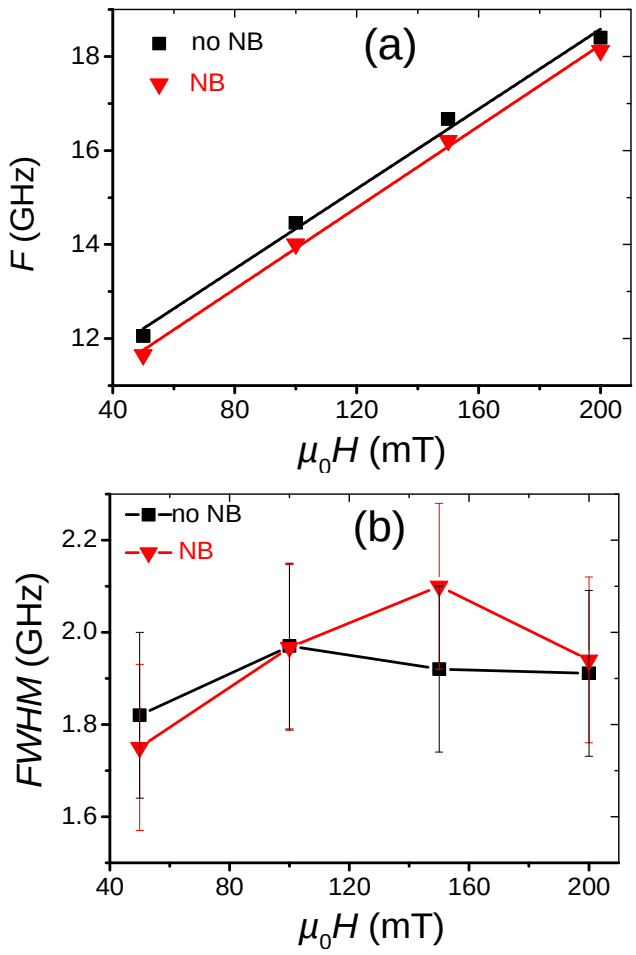

Fig. 3 (Colour online) Evolution of the magnons frequency (a) and of the linewidth (b) versus applied magnetic field. The data points acquired over the Co area are in black and over the area with NB molecules coated in red. The continuous lines are guide to eyes.

B. 1–(2-bisthienyl) benzene (BTB) measurements

We present now the results obtained with BTB molecules. In a second step we will compare them to those obtained with NB molecules, for the same experimental conditions.

Figure 4 shows typical raw BLS spectra (anti-Stokes side) for BTB coated and non-coated areas of the Co film. As for the NB molecules, the magnons are measured over both areas. One can observe that for this latter area the frequencies are lower than those measured in the Co area without BTB molecules coating. On the other hand, the experimental linewidth (*FWHM*) is measured about 2.2 GHz in the area coated with molecules while it is only about 1.2 GHz in the non-coated one, for all the investigated range of the applied magnetic field.

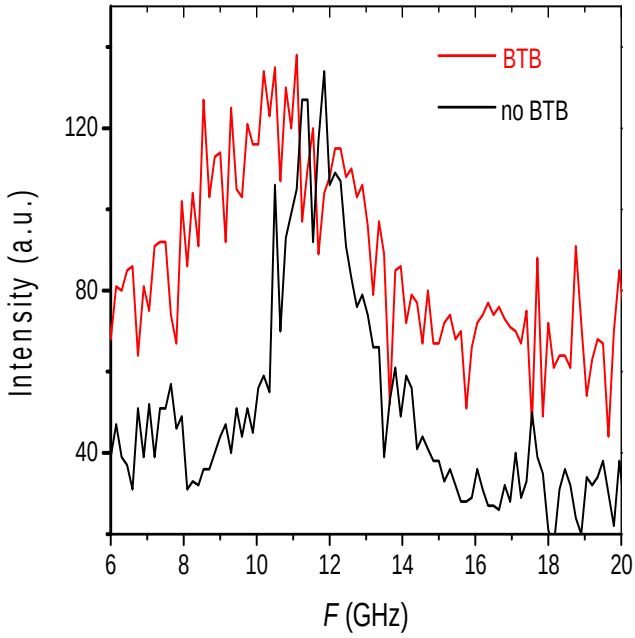

Fig. 4 (Colour online) Raw BLS spectra (anti-Stokes side) obtained under a 100 mT in plane magnetic field and a wavenumber k equal to 13.5 μm^{-1}; black: Co area (with presence of CoO); red: area coated with BTB molecules.

Figure 5 shows the evolution of the measured frequencies as function of the applied magnetic field for an angle of incidence of 35° corresponding to a wavenumber k = 13.5 μm^{-1}. We can observe that in this case the frequency shift between the two investigated areas is greater than for the NB sample, indicating a stronger effect of the coating of BTB molecules comparing to the NB molecules.

We now discuss the change of frequency in NB and BTB modified areas. First, the frequency of the pure Co magnons is different even between the two samples. At 200 mT, the frequency is about 18 GHz for the NB sample while it is only about 15 GHz for the BTB sample. The Co thin films came from different fabrication batches and thus their degree of oxidation might be different. Therefore, the two investigated Co samples present different properties. It is to note that the oxidation of the Co from oxygen of air reaches a passive state without any further evolution. Indeed, we checked that magnons peaks have the same frequency for the same sample and experimental conditions, even after several months at air.
If we consider that the change of the frequencies in the presence of molecules over the covered area is solely because of change in the Co thickness, we obtained a huge difference

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in the thickness, frequency, after coating, between NB and BTB. However as the electrochemical coating proceeds from the same diazonium electrochemistry the same change in thickness and thus frequency is expected.

As we did not observe the same frequency difference δF between coated and non-coated areas for BTB ($\delta F \approx 1.3$ GHz) and NB ($\delta F \approx 0.5$ GHz) molecules, this frequency shift can be at least partially attributed to an effective anisotropy variation. In fact, the coated samples present the same nominal thickness then the probable way to modify the effective anisotropy is a variation of the surface anisotropy. In order to analyze the observed behavior, we roughly estimated the variation of the effective anisotropy by using equation (1).

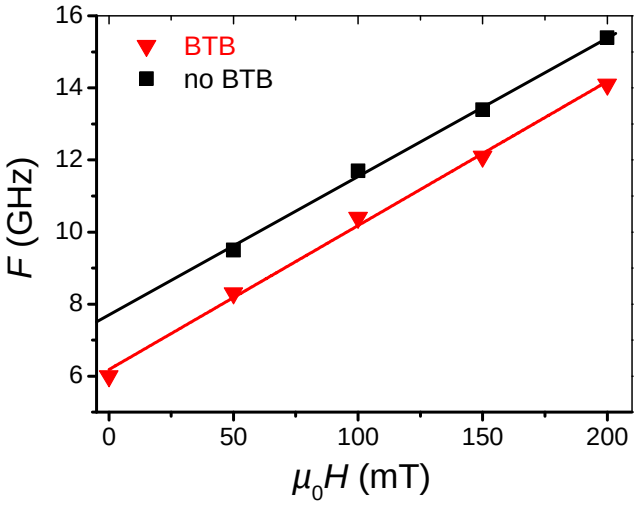

Fig. 5 (Colour online) Evolution of the magnons frequency versus applied magnetic field. The data points acquired over the Co area are in black and over the area coated with BTB molecules in red. The continuous lines are guide to eyes.

Let us first consider the case of BTB coated sample. We hypothesize that cobalt has the properties of bulk material with the following parameters: $\gamma/(2\pi) = 30$ GHz/T; $A_{ex} = 2\times10^{-11}$ J/m and $\mu_0 M_s = 1.76$ T. The measured frequency difference between the two investigated areas is $\delta F \approx 1.3$ GHz. If this frequency variation is due to a change in the anisotropy, then we obtain a variation of the anisotropy field $\delta H_K \approx 0.145$ T corresponding to a variation of the anisotropy constant $\delta K \approx 103$ kJ/m^3.

Let us now consider the case of NB coated sample. Using the above mentioned bulk parameters, from the measured frequency variation $\delta F \approx 0.5$ GHz, we obtain a variation of the anisotropy field $\delta H_K \approx 0.048$ T corresponding to a variation of the anisotropy constant $\delta K \approx 51$ kJ/m^3.

Our results are consistent with the studies reporting that organic molecules affect the surface magnetic properties mainly through interfacial effects [2, 5, 6, 10-13].

Concerning the *FWHM* linewidth results, two possibilities arise. First possibility, the change of damping is due to interfacial spin pumping modification which is unlikely due to the weak electronic density present in molecules. Second possibility, the Co magnetic properties are modified due to a change in the interfacial electronic structure due to the coupling with the molecules.

In this sense, we propose that changes in magnetization dynamics are related to the hybridization of the π-electrons of the aromatic structures with the Co surface [5, 10, 16, 17]. Indeed, it has been demonstrated theoretically that magnetic hardening or softening can be induced in the case of ferromagnetic surface interacting with nonmagnetic organic molecules containing a single π–bond [16]. Still, by means of *ab initio* calculations [10], it has been shown that the spin polarization of a ferromagnetic surface can be locally tailored by adsorbing organic molecules containing $\pi(p_z)$-electrons on it through hybridization of these out-of-plane orbitals with the spin-polarized *d* states of the metal. Moreover, this spin polarization depends on how strongly the molecules are linked to the surface. For instance, a spin polarization reversal due to the differing hybridization between Co-phthalocyanine molecules and each cobalt electrode has been evidenced in Co/CoPc/Co magnetic tunnel junctions [13]. Interface coupling was also found to strongly influence spin injection as demonstrated in the case of (La,Sr)MnO_3/Alq_3/Co magnetic tunnel junctions [1], or to stabilize an antiferromagnetic ordering at room temperature at the interface between a cobalt ferromagnetic layer and a paramagnetic organic manganese phthalocyanine (MnPc) layer [12]. Interestingly, we have observed that both change of anisotropy and increase of the damping are more important with the BTB molecule than with the NB one. Spectroscopy measurements and DFT calculations have to be performed to understand the difference in the electronic structure between those two molecules over a Co surface. The efforts must be carried forward on this effect on the magnetization dynamic aspects in order to advance towards applications in ultrahigh frequency molecular spintronic devices.

IV. CONCLUSION

We show in this work, through BLS measurements, that different organic molecules covalently coating a Co layer have different effect on magnetization anisotropy and dynamics. Two different molecules, BTB and NB, were electrocoated on Co surface through diazonium-based electrochemistry. This electrochemical coating leads to a modification of the Co film in both cases as the same chemistry mechanism is involved. However, measured magnetization properties over the coated areas depend on the molecule nature. In NB coated area, magnons show no change in the width of the BLS peaks unlike an important modification for BTB molecules coating. These results are important for the development of molecular spintronic. Further experiments are required in order to optimize the electrochemical coating procedure and then to quantify the tuning of the properties.

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