

Freestanding magnetic membranes on graphene for spin-filtering applications

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Spin polarimetry of low-energy electron beams is of considerable importance for a wide range of applications. However, an efficient method for two-dimensional, quantitative spin mapping is still lacking as state-of-art detectors rely on the sequential measurement of the spin polarization at individual points in energy and momentum space. In this work, we exploit the spin-dependent transmission of electrons through ultrathin magnetic layers embedded in a suspended matrix of a few graphene layers, fabricated in the form of micrometric magnetic freestanding membranes with an overall thickness below 10 nm, allowing significant transmission of low-energy electrons. We systematically investigate the role of deposition process, number of graphene layers, and magnetic materials with both in-plane and out-of-plane magnetization. We end up with optimized fabrication conditions for producing highly reliable elastic membranes with energy-dependent transmittivity suitable for spin filtering. We also present an analytical model that describes the detection of the spin polarization of an electron beam and outlines the experimental conditions under which such measurements can be performed using suspended magnetic membranes. This research paves the way for the development of spin filters that can be seamlessly integrated into existing detection systems, enabling spin-, angle-, and energy-resolved photoemission experiments as add-on functionality.

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I. INTRODUCTION

Measuring the spin polarization of electrons in matter is of fundamental importance in physics and in material science developments for spintronics and advanced sensors. The emergence of ferromagnetism, antiferromagnetism, and other order parameters in quantum materials depends on the energy scale of spin interactions in solids, which ranges from eV of exchange, to 10–100 meV of spin-orbit interactions, to external-field-dependent Zeeman interactions. Topological insulators [1], bulk Rashba materials [2], Weyl semimetals [3], and altermagnets [4] are examples of emerging solid systems requiring spin sensitivity in the determination of their relevant electronic properties by either spectroscopy or transport measurements. Angle-resolved photoemission spectroscopy (ARPES) gives direct access to the electron state dispersion relation $E(\mathbf{k})$ and allows for comparison with

first-principle band-structure calculations. The comparison of state-of-the-art spin-resolved experimental measurements and calculations carried out by density-functional theory is essential to interpret the results on quantum materials and systems (interfaces, nanostructures), as well as to tailor their properties to explore potential functionalities. As demonstrated in many recent advances [2,5,6], the spin orientation $\mathbf{s}$ for carriers in their electronic bands plays a key role in all these materials and needs to be efficiently detected in a photoemission experiment to obtain the spin-resolved energy dispersion $E(\mathbf{k},\mathbf{s})$.

In spin-resolved ARPES (S-ARPES), the efficient measure of the electron spin polarization (SP) remains the actual technological challenge. The SP of an electron beam cannot be separated by an external gradient of magnetic field, due to the Lorentz force. Mott polarimetry is instead based on scattering in the Russell–Saunders coupling scheme (L-S scattering) of high-energy electrons (50 keV–1 MeV) from target materials with large spin-orbit interaction, and generates a left-right asymmetry from the scattering of a single bunch of electrons polarized up or down with respect to the scattering plane [7]. In the single scattering approximation, valid for electron energies of several tens or hundreds of keV, the Mott polarimeter is “self-calibrated” as it follows a Sherman function that

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can be calculated. The technique was developed for experiments on beta decay and later for the first experiments of electron spin polarimetry of ejected electrons from solid surfaces [8–10], and is nowadays used for time-resolved magnetometry from surfaces, although its main limitation is the low efficiency, of the order of 10^{-4} . An alternative approach to obtain an imaging-type spin polarization detection is photoemission electron microscopy [11], which exploits spin-selective electron mirrors capable of analyzing many points of a two-dimensional (2D) electron distribution simultaneously: in this case the spin selectivity results from spin-orbit interaction in low-energy electron scattering on W or Ir targets, as measured in reflectivity mode. In this case, the definition of the effective Sherman function is fully empirical. Another approach exploits exchange scattering processes of electrons at valence-band energies. This leads to an increase in sensitivity by factor 100 with respect to $\mathbf{L}\text{-}\mathbf{S}$ scattering and is nowadays employed in the most efficient S-ARPES spectrometers. Very-low-energy electron diffraction (VLEED) polarimeters [12] represent the state-of-the-art in S-ARPES experimental setups [13]. They are based on the selective scattering of electrons, which depends on the relative orientation of the spin with respect to the magnetization of a ferromagnetic target [14,15]. Despite the gain of 2 orders of magnitude in efficiency with respect to high-energy Mott scattering, enabling the simultaneous exploitation of energy, momentum, and spin resolution, the polarimetry is performed either by integrating the signal over a small area of the exit plane of the electrostatic energy analyzer or integrating over a time interval of the time-of-flight detector that inevitably reduces the $E\text{-}\mathbf{k}$ resolution

with respect to standard ARPES. This also implies that each energy- and $\mathbf{k}$ -selected electron beam must be probed sequentially in order to reconstruct a multidimensional $E\text{-}\mathbf{k}$ -s dispersion, making the technique time inefficient. An attempt to overcome this issue was proposed in 2016 by Ji *et al.* [16]: they developed a multichannel VLEED polarimeter that enhances the performance of the technique moving from pointwise to bidimensional resolution in $E\text{-}\mathbf{k}$ space, thus drastically reducing the duration of the measurements. However, this solution requires a very complex and extended apparatus of electrostatic lenses.

An alternative, and radically different, approach exploits spin-dependent transmission through a spin-polarized material embedded in an ultrathin suspended film, acting as a spin filter. For decades, spin filtering by ferromagnetic thin films has been explored. Despite the efficiency of the spin filtering, the attenuation length of low-energy electrons travelling through solids is extremely short, so the signal quickly vanishes as the detector thickness increases beyond a few nanometers.

A novel implementation of the spin filter was proposed in 2017 by Övergaard *et al.* [17]. The spin filter is realized as a matrix of magnetic pixels fabricated on a silicon-on-insulator substrate through a multistep lithographic process (including deep reactive-ion etching and contact lithography). Such a filter might be interposed between the exit slits of an electrostatic hemispherical electron analyzer for ARPES and the 2D electron counter, such as a multichannel plate (MCP), a charge-coupled device (CCD), or a complementary metal-oxide semiconductor detector, collecting an interval of energy dispersion and momentum dispersion as a map [see Fig. 1(a)]. The

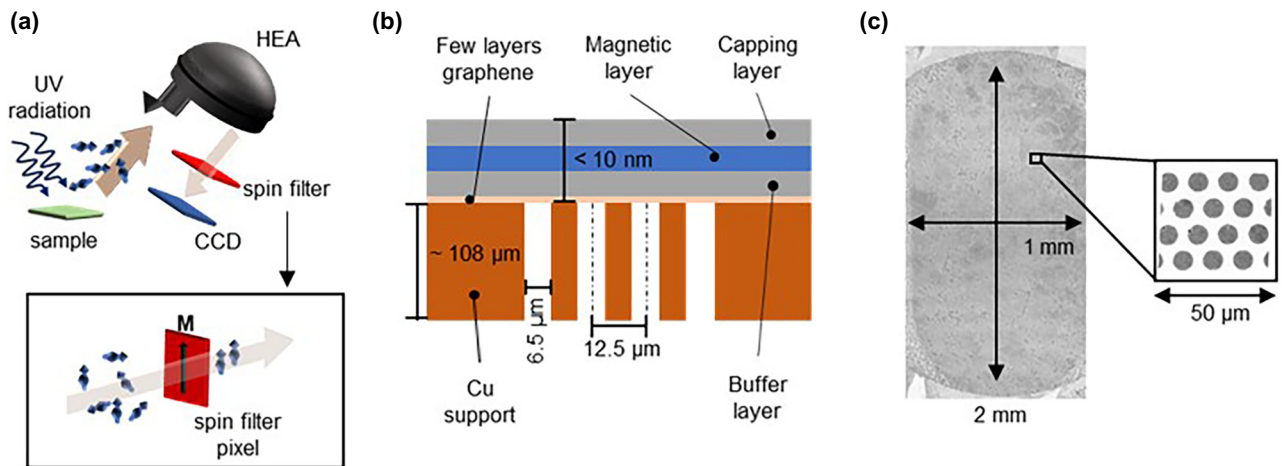


FIG. 1. (a) Schematic of a photoemission spectroscopy experiment with angle and spin resolution. Spin-filtered photoelectrons are directly collected by the charge-coupled device (CCD); HEA denotes hemispherical electron analyzer. The bottom panel is a schematic of the filtering process by a magnetic pixel with a fixed magnetization vector in the ideal case of 100% spin selectivity (only electrons with magnetic moment parallel to pixel magnetization can pass through). (b) Sketch of the cross section of the proposed structure for the suspended magnetic layer. The heterostructure is suspended on top of a copper template. (c) Scanning electron microscopy image of the 1×2 mm² surface of the filter containing freestanding membranes. The inset provides an enlarged view of a few membranes.

filter determines the different transmission of electrons depending on their spin orientation versus the quantization axis defined by the magnetization of the filter pixel [18–20], and thus a different number of electrons reaching each pixel of the CCD. This solution, exploiting the excellent performance of state-of-the-art CCDs (noise < 3 cps, linearity $> 10^6$), could ideally add a magnetization-sensitive signal per pixel when collecting energy- and $\mathbf{k}$ -dispersed photoelectrons. However, the design and construction of such a filter present technological and operational challenges.

A key work by Tereshchenko *et al.* [21] has recently demonstrated a directly integrated ferromagnet-MCP detector based on a thin Co/Pt multilayer on SiO₂, realized using a sacrificial layer of nitrocellulose. This work represents the state of the art in dedicated spin-imaging tubes. However, the monolithic integration of the spin filter with the microchannel plate (MCP) imposes intrinsic constraints: the detection is limited to a single spin component, and the filter cannot be removed to increase the count rate in experiments where spin resolution is not required.

In this work, we implement an alternative approach to building a standalone 2D spin filter to be put in front of the MCP using graphene-based freestanding magnetic membranes. In contrast with other existing polarimeters [13–16,21], our proposed polarimeter does not require a complete redesign or modifications to the detection system and can be installed as a simple add-on to the existing detection setup. Either the change of the membrane in front of the MCP or the creation of regions with different magnetization orientation allows for the detection of all the components of the spin of an incident electron beam by using membranes engineered with in-plane or out-of-plane magnetic anisotropy. The removal of the membrane allows for maximizing the counts when band dispersion is acquired, regardless of the spin. Notably, this modular filter is also characterized by a relatively simple fabrication process. Moreover, a model to quantify the transmitted current through membranes as a function of their parameters is provided.

The choice of the heterostructure allows for mechanical robustness and spin-selectivity; as sketched in Fig. 1(b), it consists of the following layers:

- (1) a metallic Cu grid with a suitable arrangement of densely packed holes to support the bidimensional matrix;
- (2) a mechanically robust layer, composed of few layers of graphene, to make a freestanding membrane corresponding to the holes of the metallic grid;
- (3) a buffer layer (made of either Au or Ru) as a seed layer for the smooth growth of the topmost magnetic layer;
- (4) an active magnetic element (made of a single layer of Co or a multilayer with CoFeB) that allows for spin filtering, with high magnetic remanence and large magnetic coercivity, as well as good mechanical stability;

- (5) a capping layer (either Au or Ru) to protect the magnetic material from contamination after air exposure.

Considering the exponential attenuation of an electron beam with the thickness of the membrane, the whole structure's thickness is limited to a few nanometers to ensure a sizeable transmission of electrons, especially for a low-energy electron beam (0–10 eV).

Here we present the deposition process of suitable magnetic stacks on freestanding graphene sheets. We investigate the membranes' intrinsic properties (morphological, elastic, magnetic), assess their mechanical reliability across different fabrication conditions (graphene thickness, deposition technique), and analyze their electron transmission despite defects partially inherited from the initial samples. An analytical framework is developed to model both transmission and spin-filtering performance, and predictions are benchmarked against experimental data. The predictive capability of the model allows for identifying the design rules (thickness, geometry, defects, etc.) and beam properties (beam shape, divergence, etc.) that determine the figure of merit of such filters.

II. METHODS

Two kinds of graphene membranes from TED Pella, Inc. were used for the realization of the spin filters. Those are composed of a copper grid on which graphene is deposited to obtain suspended membranes in the positions where the copper has holes [see Fig. 1(b)]. Graphene membranes are either two monolayers (2 ML, from now on) or 7 ± 1 monolayers (7 ML, from now on). They correspond to thicknesses of about 0.67 nm and 2.10 ± 0.34 nm, respectively (the interlayer spacing in graphite is 0.335 nm [22]). The membranes are circular, with a diameter of 6.5 μm and a center-to-center distance of 12.5 μm . The values were confirmed by imaging with scanning electron microscopy [see Fig. 1(c)].

Two sets of samples were realized by growing the magnetic heterostructures (including the buffer and the capping layers) by molecular beam epitaxy (MBE) or by magnetron sputtering to investigate the differences between the corresponding workflows and templates. Notably, several magnetic heterostructures have been investigated to achieve different directions for the magnetic easy axis and thus to provide spin selectivity to different spin components. In particular, in-plane (IP) magnetic anisotropy is obtained with Au (or Ru)(3 nm)/Co(3 nm)/Au (or Ru)(2 nm) (layers listed from bottom to top), whereas out-of-plane (OOP) magnetization is obtained by exploiting interfacial perpendicular magnetic anisotropy (PMA) in Ta(3 nm)/CoFeB(1 nm)/MgO(2 nm)/Ta(2 nm), where both the MgO and the capping layers are crucial to achieve such orientation of the magnetization, as well established in literature [23].

Heterostructures comprising cobalt were grown either by MBE or magnetron sputtering for comparison, while those containing CoFeB were exclusively grown by magnetron sputtering. Depositions by MBE were carried out in a custom ultrahigh vacuum cluster tool [24] (chamber base pressure on the order of 10^{-10} mbar) by Co and Au effusion cells at a rate of about 0.2 nm/min, as checked by means of a quartz microbalance.

For magnetron sputtering, an AJA ATC Orion 8 machine was used. Cobalt was grown in dc mode, with a base pressure of approximately 5×10^{-9} mbar and an argon pressure of approximately 6.7×10^{-3} mbar during the deposition. Films of Ru, Ta, and $\text{Co}_{0.4}\text{Fe}_{0.4}\text{B}_{0.2}$ were grown in dc mode as well, with an argon pressure of about 5×10^{-3} mbar. The deposition rates were about 2.0 nm/min for Co, 1.0 nm/min for Ru, 4.5 nm/min for Ta, and 1.5 nm/min for CoFeB, as checked by a quartz microbalance. MgO was instead deposited in rf mode at a rate of 0.4 nm/min and an argon pressure of about 3.3×10^{-3} mbar. All the processes were performed with the sample at room temperature.

The structural integrity of the membranes was inspected by scanning electron microscopy along the different steps of the fabrication process, while the topography and the mechanical properties were investigated by atomic force microscopy using a Keysight 5600LS microscope operated with standard silicon cantilevers (tip radius of 50 nm, elastic constant $k = 40$ N/m).

The magnetic characterization relied on the magneto-optical Faraday effect using a linearly polarized red laser ($\lambda = 635$ nm) modulated by a photoelastic modulator at 50 kHz.

Electron transmission was measured by exploiting the custom electron gun described in Ref. [25]. A collimated Gaussian beam, with a current of about 10 μA and a spot size < 1.5 mm in diameter at electron kinetic energy $E > 12$ eV, impinges on the array of membranes at variable energy (from 5 to 25 V). The current transmitted through the membrane is collected by a Faraday cup kept at 28 V. For spin-resolved experiments, a spin-polarized beam with a nominal polarization of 25% is achieved by exploiting electron photoemission from a GaAs photocathode in conditions of negative electron affinity, excited by a laser slightly above the resonance with the GaAs direct gap (823 nm).

III. RESULTS

The results are articulated in five sections.

In the first two sections, scanning electron microscopy (SEM) and atomic force microscopy (AFM) are used to assess the process yield (in terms of the fraction of intact freestanding membranes that are fabricated), topographical properties (such as the roughness), and elastic properties (including the breaking load) of membranes

after deposition. These sections focus on Co-based samples with in-plane magnetization, as they can be grown by both molecular beam epitaxy and magnetron sputtering, enabling a direct comparison between the two deposition techniques.

The third section addresses the magnetic properties (anisotropy and coercivity) for both in-plane and out-of-plane magnetization.

The fourth and fifth sections present the electron transmission results for Co-based filters, introducing a simple model to simulate transmission and spin filtering, and comparing the predictions with experimental data.

A. Mechanical reliability

The ultralow thickness of membranes allows for a significant transmission of electrons, although it imposes a careful study of the mechanical robustness of membranes along the fabrication workflow. Here we analyze the fraction of intact membranes through imaging by scanning electron microscopy.

We established the protocol sketched in Fig. 2(a) with two intermediate steps of sample imaging to recognize the impact of each phase of the process. We distinguish between three different cases: (1) broken graphene membranes already present on the sample as it is extracted from the shipping box; (2) damage connected to the handling of the samples during the workflow, and (3) damage induced by the deposition process. First, a pristine sample with a matrix of a few suspended layers of graphene membranes (as extracted from the box and containing an intrinsic number of defective membranes, with defects that we will refer to as *pristine damage*) is mounted on the SEM holder and undergoes a predeposition SEM imaging (*predeposition damage*). Then, the sample is transferred either on the MBE or the magnetron sputtering holder and is manipulated for the deposition of the heterostructure (operation leading to damage that we label as *postdeposition damage*). Finally, the samples undergo postdeposition imaging, and we compute the fraction of intact membranes at each step.

We consider a set of four samples as a function of two parameters: (1) the thickness of graphene membranes (2 ML vs 7 ML) and (2) the deposition technique (MBE vs magnetron sputtering). The four combinations are reported in Table I. Data from about 30 samples (almost equally distributed between the possible combinations) were collected. On average, we checked 1700 membranes per sample before and after deposition.

Figure 2(b) shows the percentage of intact membranes for the pristine samples with 2 and 7 ML of graphene ($87\% \pm 10\%$ and $94\% \pm 9\%$, respectively) with a significantly higher percentage in the case of the thicker graphene. Apart from the damage that occurred during the fabrication process followed by the producer, about

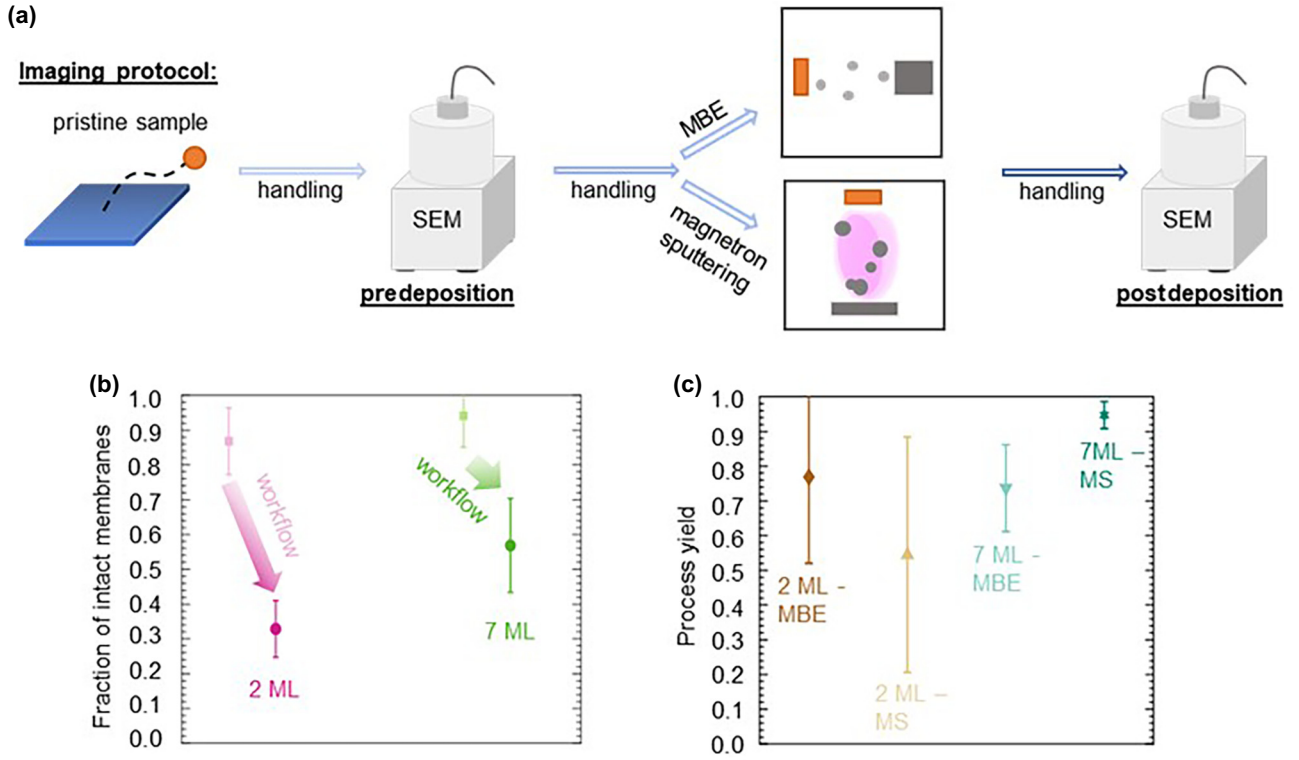


FIG. 2. (a) Sketch of the workflow with a double check by scanning electron microscopy (SEM) to disentangle deposition and sample handling as possible sources of membrane breaking. (b) Fraction of intact membranes before and after the entire workflow in the case of two monolayers (2 ML) and seven monolayers (7 ML) of graphene. (c) Process yield for the four combinations of the parameters [2 ML vs 7 ML, molecular beam epitaxy (MBE) vs magnetron sputtering (MS)]. The error bar is the standard deviation of the process yield.

which we are blind (being a commercial product), the handling of the devices during mounting would more easily damage thinner layers than thicker ones (we are considering pristine and predeposition damage, as formerly defined). Figure 2(b) shows additional damage as a result of the deposition process after which we count $33\% \pm 8\%$ and $57\% \pm 14\%$ for 2 and 7 ML of graphene, respectively. To summarize the effect of the deposition, we report in Fig. 2(c) the *deposition process yield*, defined as the ratio between the number of intact membranes post- and predeposition. The sputtering on 7 ML of graphene allows for the best process yield with a reduced variability ($95\% \pm 4\%$ of the previously intact membranes remain unaffected by the deposition process), which would ensure

TABLE I. Sample naming vs parameters (graphene thickness and deposition technique). The number refers to the graphene MLs, while the letters indicate the deposition method.

Graphene ML number	Deposition technique	
	MBE	Magnetron sputtering
2 ML	2ML-MBE	2ML-MS
7 ML	7ML-MBE	7ML-MS

quite good reliability in the production of filters. We conclude that thicker graphene (e.g., 7 ML) is desirable to guarantee a more pronounced mechanical reliability.

Further investigations into the nature of the damage were carried out to understand the impact on the electron transmission of the filters. We preliminarily state a definition of what we consider a broken membrane. SEM images shown in Figs. 3(a)–3(c) report the three possible scenarios. Figure 3(a) shows a fully intact membrane, Fig. 3(b) gives an example of an almost fully intact membrane with some small holes but a significant portion of intact area (98.5% in the specific case), and Fig. 3(c) shows a broken membrane with more than half of its area lifted off. Concerning filtering, membranes such as in Fig. 3(c) let a significant fraction of unfiltered electrons to pass without any spin filtering, which is detrimental for applications. Figure 3(d) maps the overall transmission of a membrane as a function of the damaged area (vertical axis) and the effective membrane transmission (horizontal axis). The latter can be approximated by the following heuristic expression:

$$t(a, x) = a + e^{-x}(1 - a), \quad (1)$$

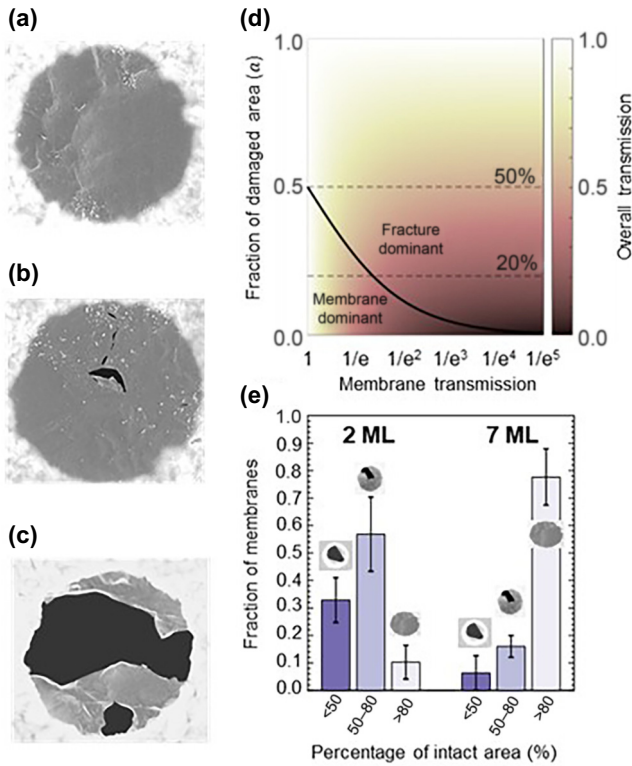


FIG. 3. Images of a graphene membrane (a) fully intact, (b) with small fractures, and (c) broken (more than half the area is damaged). (d) Color map of the overall transmission of the suspended membrane, considering the contribution of both damaged and intact membranes. The solid line indicates the boundary between the region where the transmission is dominated by the membrane and the one dominated by the damage. (e) Extent of the damage for membranes with two monolayers (2 ML) and seven monolayers (7 ML) after the deposition process.

where x is the attenuation factor and a is the fraction of the damaged membrane area. The solid black line indicates the boundary between the region of the plot in which the overall transmission is dominated by the presence of holes [first term in Eq. (1)] and the region in which the contribution of the membrane is more sizeable [second term in Eq. (1)]. We set two arbitrary thresholds on the overall transmission, corresponding to percentages of the damaged area of the membranes. Above 50% damaged area, the transmission is considered dominated by the fracture, so the membrane is no longer exploitable as a filter (and the corresponding pixel on the CCD must be deactivated). Below the threshold of 20% damaged area, the membrane selectivity is considered large enough for filtering applications (and the corresponding pixel on the CCD is enabled).

Figure 3(e) presents the fraction of membranes as a function of the intact area. It can be noticed that the damage is more severe for membranes with 2 ML of graphene, with 33% of membranes completely broken (damage >50% area, i.e., intact area <50%), 57% of partially broken

(damage between 20% and 50% area, i.e., intact area between 80% and 50%), and only 10% with small fractures (damage <20% area, i.e., intact area >80%), while for membranes with 7 ML of graphene the situation is opposite, with less than 7% of completely broken membranes, 16% of partially broken ones, and up to 78% with small fractures.

Thus, considering membranes with a relatively small fraction of damaged area as good filters, these results show that the mechanical reliability is even better than that reported in the former paragraphs [see Fig. 2(c)]. Indeed, in terms of the effective broken area (the ratio of broken area to total area) the damage reported is reduced, especially for substrates with 7 ML of graphene.

We note that the main scope of this mechanical reliability study is to quantify the impact of manipulation and deposition on the membrane integrity. We are confident that proper engineering of membrane production, handling, and magnetic stack deposition (perhaps in an automated fashion) could allow the drastic reduction in or even elimination of defective pixels.

B. Morphology and elastic properties

Atomic force microscopy was used to determine the topography and roughness of membranes as well as to investigate their mechanical properties. Figure 4(a) reports the topography of a membrane grown by MBE on a 7-ML-thick graphene layer (sample 7ML-MBE) and unveils a better surface (with a rms roughness of about 0.5 nm) than that obtained by magnetron sputtering, which presents a larger roughness (1.3 nm). From the profile in Fig. 4(a), the freestanding layer turns out to be not perfectly stretched but somewhat rippled. The bending is typical of such membranes, as shown in Ref. [22]. The maximum tilting of the surface of the membrane with respect to the horizontal plane never exceeds 1.5° and can be considered to have a negligible impact.

The mechanical properties were tested by indenting the surface of the membrane at its center using the tip of the atomic force microscope in contact mode, as sketched in the inset of Fig. 4(b). The membrane deflection was measured by subtracting the tip deflection from the piezo displacement, the former being calibrated using the rigid copper support, following the method reported in Ref. [22].

When the membrane is pushed with a relatively low force, its response is elastic with a linear displacement-force curve without hysteresis [Fig. 4(b)]. The force can be increased up to the breaking load that provokes the fracture of the membrane, detected as a steep drop in the AFM signal [marked with arrows in Fig. 4(b)]. The breaking load for sputtered graphite/Ru(3 nm)/Co(3 nm)/Ru(2 nm) membranes with 2 ML of graphene is about $1.3 \mu\text{N}$, while this value increases to about $7.3 \mu\text{N}$ ($\pm 50 \text{ nN}$) in the case of 7 ML. These results are compatible with previous studies

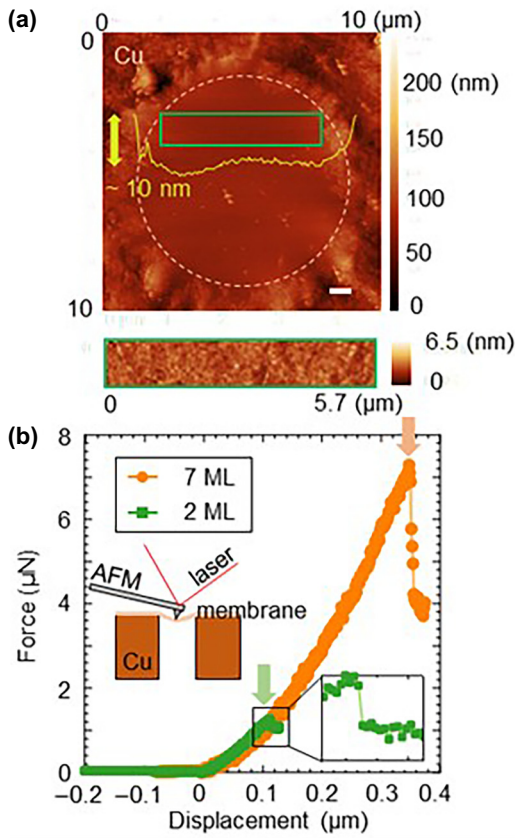


FIG. 4. (a) Atomic force microscopy (AFM) topography of a membrane grown by molecular beam epitaxy (MBE) on a seven-monolayers (7-ML)-thick graphene layer. The freestanding region is inside the dashed circle, and the AFM profile of the suspended region measured along one diameter is superimposed in yellow. The bottom inset shows the detail of the green rectangular region. (b) Force vs displacement for samples with two monolayers (2 ML; green) and 7 ML (orange) with the same Ru(3 nm)/Co(3 nm)/Ru(2 nm) heterostructure grown by MBE on top. The arrows indicate the breaking load of the membranes. The inset shows a sketch of the indentation process of the tip.

on graphene monolayers [22], supporting the idea that a few layers of graphene sheet guarantee robustness to the whole floating heterostructure.

C. Magnetic properties

Spin polarimetry requires high magnetic coercivity to guarantee the stability of the magnetization against external perturbations and temperature, as well as large remanence for a larger efficiency. Moreover, the ability to achieve both IP and OOP magnetic anisotropy enables sensitivity to the different spin components of an impinging electron beam.

Rigid samples with the same magnetic heterostructures grown on graphene were characterized by vibrating sample magnetometry (VSM). Stacks were deposited on Si(001) substrates, then capped with 1 μm of thermal

SiO₂ and patterned in the form of micrometric pillars with a lateral size of 50 μm , mimicking the template of the suspended samples. These samples aimed to confirm that, on ideal flat surfaces such as that of SiO₂/Si, the magnetic templates Au(3 nm)/Co(3 nm)/Au(2 nm) and Ta(3 nm)/CoFeB(1 nm)/MgO(2 nm)/Ta(2 nm) are magnetized IP and OOP, respectively, without asserting the same coercivity or squareness. Thicker cobalt (3 nm) is in-plane magnetized thanks to the predominance of the shape anisotropy. Thinner CoFeB (1 nm) with asymmetric interfaces (a heavy metal on one side and an oxide on the other) allows for interfacial magnetic anisotropy [23], with the key role of the topmost interface CoFeB/MgO/Ta rather than the bottom one. Importantly, the first layer of either Au(3 nm) or Ta(3 nm) is intended to decouple the substrate from the heterostructure and serves as a seed layer for the smooth growth of the topmost magnetic layer.

The direct characterization of freestanding films was instead performed by the Faraday effect [setup in Fig. 5(a)], analyzing the rotation acquired by linearly polarized laser light with a 635-nm wavelength passing through the magnetic layers. Figure 5(b) shows the comparison between the VSM and Faraday measurements for the Co-based heterostructure [Au(3 nm)/Co(3 nm)/Au(2 nm)]. The VSM characterization of the IP [see Fig. 5(b), solid line] and OOP (data not shown) magnetization components indicate an in-plane magnetic easy plane, as expected for such a heterostructure due to shape anisotropy. The IP coercive field and the squareness of the loop (the ratio between remanent and saturation magnetizations) are 8.0 mT and 0.77, respectively, on the rigid substrate (VSM, solid line). The Faraday effect (red dots) was measured by applying the magnetic field parallel to the sample and making the light impinge at $\theta = 17^\circ$ with respect to the sample normal: with such an arrangement, the projection of the magnetic moment along the direction of the laser beam is equal to $M \sin \theta = 0.29M$, where M is the in-plane magnetization. The coercive field is 3.3 ± 0.1 mT, while the normalized remanence is 0.66. Note that the former decreases and the loop sharpens with respect to the VSM measurement, possibly reflecting the role of the substrate.

Heterostructures based on CoFeB [Ta(3 nm)/CoFeB(1 nm)/MgO(2 nm)/Ta(2 nm)] were designed to achieve out-of-plane anisotropy thanks to the presence of the interfaces with Ta and MgO, which can provide PMA contributions [23]. The magnetic characterization of these membranes and the rigid counterparts are presented in Fig. 5(c). In contrast with the Co-based samples, such a heterostructure shows an easy axis in the direction perpendicular to the film [see Fig. 5(c), solid line], unveiling the expected PMA, with a hard axis in the film plane (data not shown).

The coercive field and the normalized remanence by VSM in the OOP direction are 2.8 mT and 0.72, respectively. The magnetic properties were measured by the

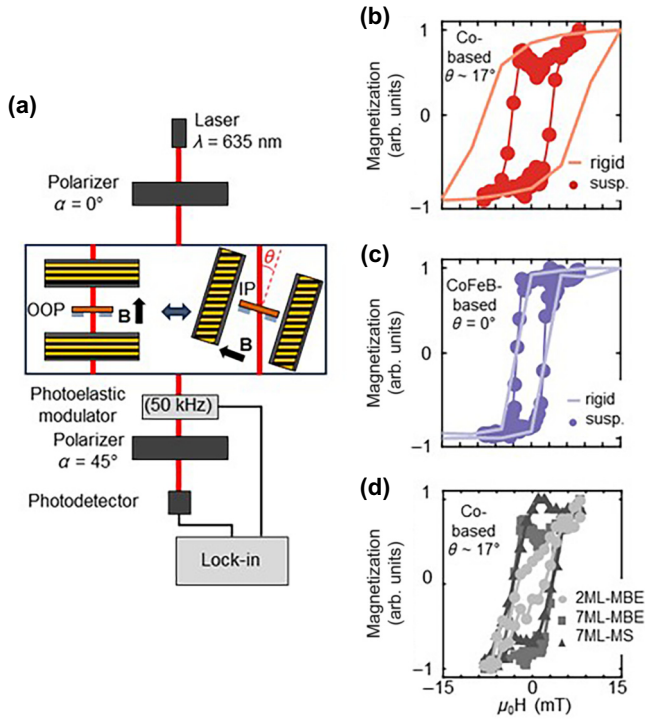


FIG. 5. (a) Faraday effect setup in in-plane (IP) and out-of-plane (OOP) configurations. (b),(c) Hysteresis loops for the IP and OOP magnetized heterostructure based on Co and CoFeB, respectively. The Faraday effect on suspended membranes (dots) is compared with vibrating sample magnetometry data from the same structure deposited on Si/SiO₂ (solid lines); (d) comparison between samples 2ML-MBE, 7ML-MBE, and 7ML-MS (ML, monolayer; MBE, molecular beam epitaxy; MS, magnetron sputtering).

Faraday effect working with the beam at normal incidence on 7-ML-thick graphene layers. Figure 5(c) shows that the magnetic behavior is similar on suspended samples: the magnetic easy axis remains perpendicular to the plane, while the coercivity and normalized remanence are 2.3 ± 0.24 mT and 0.84, respectively. By comparison with Fig. 5(b), we note that a larger difference between coercive fields on suspended and rigid samples emerges in the IP case (3.3 mT vs 8.0 mT) than in the OOP one (2.3 mT vs 2.8 mT). The OOP hysteresis loops of Fig. 5(d) are pretty much independent on the substrate, highlighting the key role of MgO/Ta in establishing perpendicular magnetic anisotropy [23] and the decoupling effect of the Ta seed layer. On the other hand, Fig. 5(b) shows that a clear difference between coercive fields on suspended and rigid samples emerges in the IP case (3.3 mT vs 8.0 mT). We ascribe this difference to the fact that the IP heterostructure on SiO₂ was patterned in the form of micrometric pillars with a lateral size about 8 times larger than the membranes, and to the influence of the bottom Au morphology on Co [26], mediated by the low wettability of SiO₂ by Au. Note

also that the coercive fields are sufficiently large to guarantee that the magnetic moments are not perturbed in spin polarimetry setups, which are typically already magnetic-field-free in order to avoid any deflection in the electron trajectories (as a comparison, it is noteworthy to cite the case of the state-of-art VLEED detector based on Fe(001)-*p*(1 × 1)O targets, which presents coercive fields of just 0.1 mT [14] and is thus more sensitive to any external spurious magnetic field). Additionally, the beam polarization is not expected to give any perturbation on the membrane magnetization.

Note that, to correctly measure the beam polarization, it is necessary to compute the difference between the transmitted signals through membranes with opposite magnetizations. We define this difference $\Delta I = (I_+ - I_-)$, where I_+ (I_-) is the transmitted current when the net beam polarization, before the membrane, is spin up (down), i.e., antiparallel (parallel) to the membrane magnetization. A single membrane can be used for this scope, reversing the magnetization continuously with an external magnetic field generated by a coil and exceeding the magnetic coercivity. Otherwise, two different membranes magnetized with opposite magnetizations can be moved alternately in front of the MCP. The second solution would avoid the generation of magnetic fields in the inner space of the hemispherical analyzer, eliminating the risk of modifying the electrons' trajectories by spurious local magnetizations.

Having assessed the IP (OOP) magnetic anisotropy of the Co (CoFeB) samples, we further investigated the role of the buffer and capping layers by comparing samples 2ML-MBE, 7ML-MBE, and 7ML-MS with the IP magnetic configuration, as shown in Fig. 5(d). The main features are maintained. Coercive fields and remanence states are comparable, especially in thicker samples. The coercive field is around 3 mT for all the samples. The 2ML-MBE membrane exhibits an elongated hysteresis loop with relatively low remanence (approximately 50% of the saturation magnetization). We believe that this behavior is due to the higher proportion of damaged membranes in this sample [around 60% of the total, see Fig. 3(e)], which likely deteriorates the average magnetic response as a result of boundary effects, size effects, mechanical bending, and other issues associated with broken membranes. In contrast, membranes grown on thicker graphene layers (7 ML) are generally less fragile and remain intact in most cases [approximately 80% of the total, see Fig. 3(e)], resulting in squared magnetic hysteresis loops. Only minor differences are observed between MBE- and sputter-deposited membranes in the 7 ML case.

D. Transmission

Transmission experiments were carried out employing the electron gun described in Ref. [25]. The setup is

sketched in Fig. 6(a). Electrons are photoemitted from a GaAs photocathode, illuminated by a near-infrared laser diode with 823-nm wavelength in conditions of negative electron affinity. They pass through a cylindrical capacitor that rotates the beam propagation direction by 90°. In this condition, properly optimized electron optics focus the beam on the membrane with normal incidence and spin parallel to the sample surface. The membrane partially transmits the beam to a Faraday cup on the back, where its intensity is measured. The transmission was first detected on bare graphene membranes (7 ML) and then after depositing 1.5 nm and 3.5 nm of cobalt to investigate the role of the magnetic layer in in-plane magnetized samples. These were used as a test bench for characterizing electron transmission through the graphene-based membranes with minimal thickness to maximize the signal-to-noise ratio. OOP samples were not used due to their greater thickness and the need to retain the full CoFeB/MgO/Ta trilayer to preserve magnetic anisotropy. Co was deposited *in situ* in the conditions reported in Sec. II, without any buffer layer to concentrate on the role of the magnetic material, avoiding any additional contribution. The latter case (graphene 7 ML + Co 3.5 nm) is very close to sample 7ML-MBE, where 3 nm of Co was used. All the measurements were performed at room temperature.

Membranes are grounded thanks to direct contact with the Cu grid support so that any charging effect that would modify the beam spread and/or energy is excluded.

Here we model the current transmission of the electron beam through the filter taking into consideration three main contributions.

First, the transmission of electrons through intact membranes is attenuated according to the Beer-Lambert law,

$$T(t, E) = \exp(-t/\lambda_{\text{IMFP}}), \quad (2)$$

where t is the thickness of the suspended membrane and λ_{IMFP} the electron inelastic mean free path, calculated from its universal function $\lambda_{\text{IMFP}} = 143E^{-2} + 0.054\sqrt{E}$ [27]. The energy E is defined as the effective kinetic energy seen by the membrane,

$$E = eV_S + E_{\text{gap}} - \phi, \quad (3)$$

where E_{gap} is the GaAs photocathode energy gap (1.42 eV at room temperature), e is the electron charge, V_S is the membrane potential [see Fig. 6(a)], which can be varied between 0 and 20 eV, and ϕ is the membrane surface barrier potential, which we assumed to be coincident with the work function (4.23 eV for graphene [28] and 5 eV for Co [29]).

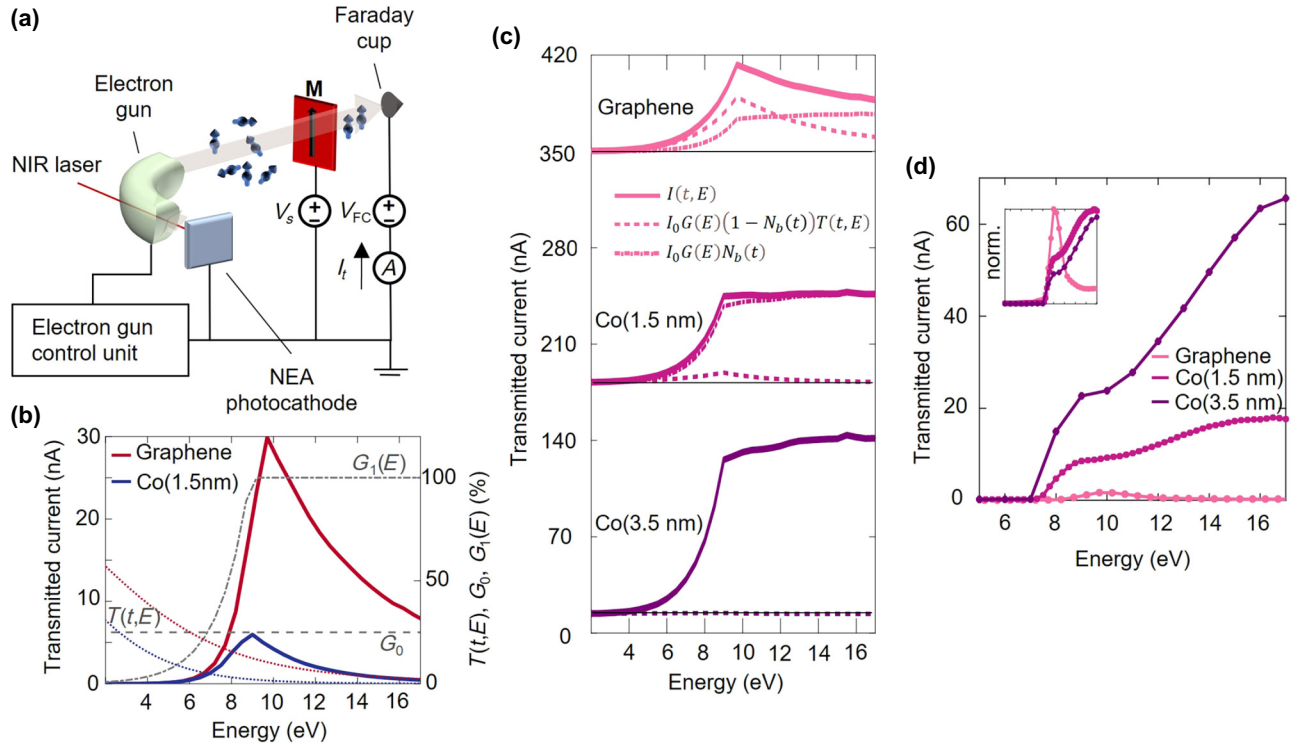


FIG. 6. (a) Experimental setup used to study the filters using an electron gun (NIR, near-infrared; NEA, negative electron affinity). (b) Transmitted current across the device, assuming all membranes are intact [Eq. (5) of the text]. (c) Calculated transmitted current vs energy for the three devices considered, assuming that a portion of the membranes is broken [Eq. (6) of the text]. Dashed and dash-dotted lines represent the unbroken and broken membrane contributions. Data are vertically shifted by an arbitrary constant for a clearer visualization. (d) Experimental transmitted current for the three devices (normalized in the inset).

In the energy range considered here (2–17 eV), λ_{IMFP} monotonically decreases with E (being dominated by the first term proportional to E^{-2}) and so does $T(t, E)$, as shown in Fig. 6(b) (red and blue dotted curves for bare graphene and Co-capped membranes, respectively).

The second contribution to the current transmission originates from a geometrical effect, i.e., the ratio between the area of the membrane intersected by the beam and the total beam area. This term can be written as

$$G(E) = G_0 G_1(E). \quad (4)$$

G_0 is a constant and represents the ratio between the active area of the filter (the membranes only) and the total area of the device [membranes and Cu support grid, see the inset of Fig. 1(c)]. Its value is around 0.25, which means that suspended membranes cover 25% of the total area of the filter. The term $G_1(E)$ takes into account the energy-dependent spot size of the beam, which varies remarkably as the beam energy decreases and influences the number of electrons impinging on the active area of the filter and giving rise to the effective transmission. Below $E \sim 6.5$ eV [corresponding $V_S \sim 10$ V in Ref. [25], according to Eq. (3)] the beam spot size exceeds 2 mm, so that it impinges not only on the active area of the filter with the membranes [Fig. 1(c)], but also on the perimetral copper grid support that does not contribute to the output current. We considered a Gaussian beam profile with the spot size dimension as a function of the energy, reported in Ref. [25], and we evaluated G_1 as the ratio between the beam intensity inside the 1×2 mm² filter area [see Fig. 1(c)] and the total beam intensity. G_1 displays an inversely proportional behavior of the beam size versus the energy up to a saturation point, achieved at $E \sim 9.2$ eV, where the beam stays fully inside the filter area.

In Fig. 6(b) we plot the geometrical terms, G_0 (dashed horizontal line) and G_1 (dash-dotted line). Note that G_0 and G_1 are independent of the sample structure, while the transmission $T(t, E)$ depends on the total thickness t of the membrane.

The overall transmission of the 1×2 mm² filter area, assuming 100% of membranes are intact, is given by

$$T_A(t, E) = T(t, E)G(E) = T(t, E)G_0 G_1(E) \quad (5)$$

and is reported in Fig. 6(b) for bare graphene and Co-capped membranes (red and blue lines, respectively). Note that the nonmonotonic behavior, with a sharp peak around 9–10 eV, arises from two opposite contributions: $T(t, E)$, expressed by Eq. (2), which decreases when E increases because of the λ_{IMFP} dependence on E ; $G(E)$, expressed by Eq. (4), which increases with E because of the G_1 dependence on E .

The third contribution to the current transmission of the overall filter is given by the presence of holes in the

broken membranes, which significantly influence the final current measured by the Faraday cup. As a matter of fact, holes do not filter the electrons according to Eq. (2) but leave the beam to pass unattenuated, adding a spurious spin-independent current impinging on the Faraday cup. This contribution was modeled by defining a thickness-dependent fraction of broken membranes on their total number, named $N_b(t)$, exploiting the experimental data reported in the previous section. It is worth noticing that, to simplify the model, no distinctions are made between small or large fractures: a membrane is considered intact if no holes are present; otherwise, it is considered broken.

Hence, the transmitted current $I(t, E)$, impinging on the Faraday cup, reads as follows:

$$I(t, E) = I_0[1 - N_b(t)]T_A(t, E) + I_0 N_b(t)G(E), \quad (6)$$

where I_0 is the electron beam current before the device.

Note that both terms are proportional to the geometrical contribution $G(E)$, due to the ratio between the membrane area effectively illuminated and the total beam area [see Eq. (4)], so that Eq. (6) can be rewritten as

$$I(t, E) = I_0 G(E) \{ [1 - N_b(t)]T(t, E) + N_b(t) \}. \quad (7)$$

While the first term [proportional to the fraction of intact membranes, $1 - N_b(t)$] is solely due to membranes through which the beam is transmitted with electron transmission $T(t, E)$, the second one [proportional to the fraction of broken membranes, $N_b(t)$] represents the spurious contribution caused by the presence of holes, and no attenuation of the beam takes place in this case. Using the average values previously obtained for $N_b(t)$ (5.9%, 14%, and 28% for the uncapped graphene, the 1.5-nm-thick Co, and the 3.5-nm-thick Co overlayers, respectively) and the experimental I_0 current (measured sending the beam directly onto the Faraday cup without any sample between) as inputs for the model, the transmitted current calculated by Eq. (7) is plotted in Fig. 6(c) for the three steps. The dashed and dash-dotted lines represent the first and the second terms of Eq. (6), ascribed to intact and broken membranes, respectively. As expected from the model, the presence of holes makes the overall transmission increase when the thickness of the Co layer increases because the number of holes increases too. In this way, the first contribution of Eq. (6) for the 3.5-nm-thick Co sample is almost zero because of the large attenuation due to the sample thickness, so the signal arises almost entirely from the contribution of the holes.

The experimental transmitted currents, measured by the Faraday cup, are reported in Fig. 6(d). Qualitatively, data agree with the prediction of the theoretical model expressed by Eq. (7), as better evidenced by the normalized curve in the inset. The strong attenuation of the signal at small E can be attributed to the electron beam divergence, which spreads the beam on an area far larger than

the device itself [$1 \times 2 \text{ mm}^2$, see Fig. 1(c)], in accordance with the geometrical term $G_1(E)$ reported in Fig. 6(b). At larger energies, it is possible to observe the two contributions of intact and broken membranes, as discussed above. On bare graphene, as the beam energy E is increased, the transmission reaches a maximum below 10 eV whereas then decreases, and when Co is added the signal increases monotonically. A saturation value is achieved at about 19 eV for the thinner Co layer, while for the thicker layer saturation can be observed at energies greater than 20 eV, beyond the probed energy range.

Note that our model considers an electron beam normally incident on the membrane, with all electron trajectories parallel. However, due to the Liouville theorem, an angular spreading of the electron trajectories takes place on a solid angle inversely proportional to the beam energy (the beam divergence has been measured to be $>15^\circ$ at $E < 4 \text{ eV}$ and less than 2° at $E > 12 \text{ eV}$, see Ref. [25]). Owing to the device geometry [see Fig. 1(b)], all electrons crossing the membrane with an incidence angle with respect to the sample normal larger than 3.4° will end their trajectory on the vertical sides of the Cu (see also Appendix A). To have at least 50% (90%) of the electrons not colliding with the Cu grid, the incidence angle must be smaller than 1.7° (0.3°). The probability of a transmitted electron passing the Cu grid thus sharply increases with the beam energy: this observation can reasonably explain the experimental transmission onset and saturation point moving to much larger energies than the simulated ones, where this effect is not accounted for.

Apart from this side effect, notably, we observe that our heuristic model fully justifies the experimentally observed trend and quantifies the role of the holes in determining the current after crossing the filter, which should correspond to the current impinging on the detector in an actual ARPES experiment.

IV. DISCUSSION

Finally, spin-resolved experiments were performed using a spin-polarized electron beam, with the spin direction parallel to the sample in-plane magnetization, obtained using the same electron source employed for the transmission characterization originally developed and used for spin-resolved inverse photoemission experiments [25,30–32].

To evaluate the expected current asymmetry, we first model the spin-filtering process and then we discuss the transmitted current asymmetry that could be obtained in different operative conditions. Finally, we comment on the application of the proposed setup to electron spin polarimetry experiments of monochromatic and non-monochromatic beams.

A. Spin-filtering model

We start by considering the beam before crossing the membrane. The total beam intensity can be written as $I_0 = I_{0\uparrow} + I_{0\downarrow}$, where $I_{0\uparrow}$ ($I_{0\downarrow}$) is the spin-up (spin-down) component of the beam. The relation between the two components is given by the beam polarization,

$$P = \frac{I_{0\uparrow} - I_{0\downarrow}}{I_{0\uparrow} + I_{0\downarrow}}. \quad (8)$$

Our electron gun produces a beam with an absolute polarization $|P| = 0.25$ [25]. A net spin-up beam polarization (+) corresponds to $I_{0\uparrow} > I_{0\downarrow}$, that is $P > 0$ (or $P = |P|$), while a net spin-down beam polarization (−) corresponds to $I_{0\uparrow} < I_{0\downarrow}$, that is $P < 0$ (or $P = -|P|$).

From Eq. (8) and $I_0 = I_{0\uparrow} + I_{0\downarrow}$, we can obtain the two components as a function of P and I_0 ,

$$I_{0\uparrow} = \frac{1 + P}{2} I_0. \quad (9)$$

$$I_{0\downarrow} = \frac{1 - P}{2} I_0. \quad (10)$$

The transmitted beam current asymmetry, measured by the Faraday cup after crossing the membrane, is the quantity that gives the experimental contrast between the two spin polarization components after the membrane. We defined it in Sec. III as $\Delta I = (I_+ - I_-)$, where I_+ (I_-) is the transmitted current when the net beam polarization, before the membrane, is spin-up (spin-down), i.e., antiparallel (parallel) to the membrane magnetization.

The transmitted beam asymmetry is defined as the current asymmetry normalized to the total current,

$$A = \frac{I_+ - I_-}{I_+ + I_-}. \quad (11)$$

The spin-up and spin-down components of the electron beam after the membrane can be described using Eq. (6), extended to include spin-dependent inelastic mean free paths in the Beer-Lambert framework,

$$I_{\uparrow} = I_{0\uparrow} G(1 - N_b)T_{\uparrow} + I_{0\uparrow} G N_b, \quad (12)$$

$$I_{\downarrow} = I_{0\downarrow} G(1 - N_b)T_{\downarrow} + I_{0\downarrow} G N_b. \quad (13)$$

The total transmitted current ($I = I_{\uparrow} + I_{\downarrow}$) for a given beam polarization P is the sum of the spin-up and spin-down terms,

$$I = G(1 - N_b) \left[\frac{T_{\uparrow} + T_{\downarrow}}{2} + P \frac{T_{\uparrow} - T_{\downarrow}}{2} \right] I_0 + G N_b I_0. \quad (14)$$

Let us separately consider the $+$ and $-$ cases, corresponding to $P = |P|$ and $P = -|P|$, respectively,

$$I_+ = G(1 - N_b) \left(\frac{T_\uparrow + T_\downarrow}{2} + |P| \frac{T_\uparrow - T_\downarrow}{2} \right) I_0 + GN_b I_0, \quad (15)$$

$$I_- = G(1 - N_b) \left(\frac{T_\uparrow + T_\downarrow}{2} - |P| \frac{T_\uparrow - T_\downarrow}{2} \right) I_0 + GN_b I_0. \quad (16)$$

The transmitted beam asymmetry is, from Eq. (11),

$$A = \frac{(1 - N_b)|P|(T_\uparrow - T_\downarrow)}{(1 - N_b)(T_\uparrow + T_\downarrow) + 2N_b}. \quad (17)$$

For an ideal fully intact filter ($N_b = 0$), we would have

$$A = |P| \frac{T_\uparrow - T_\downarrow}{T_\uparrow + T_\downarrow} = |P|S, \quad (18)$$

where

$$S = \frac{T_\uparrow - T_\downarrow}{T_\uparrow + T_\downarrow} \quad (19)$$

is the Sherman function, which quantifies how successfully the polarimeter (in our case, the membrane) can discern between opposite spin polarizations. Note that the difference between $T_\uparrow$ and $T_\downarrow$ may arise from multiple factors, such as the spin dependence of the inelastic mean free path (i.e., $\lambda_{\text{IMFP},\uparrow} \neq \lambda_{\text{IMFP},\downarrow}$) and the spin-dependent availability of electronic states in the Co layer. A detailed calculation of these effects lies beyond the scope of this work; therefore, we rely on the experimentally determined Sherman function from Ref. [17].

Using the definition of S , the transmitted beam asymmetry [Eq. (17)] can finally be rewritten as

$$A = \frac{|P|S}{1 + [N_b/(1 - N_b)T]}, \quad (20)$$

where we define

$$T = \frac{T_\uparrow + T_\downarrow}{2} \quad (21)$$

as the transmission coefficient of an unpolarized beam.

Note that both the values of S and spin polarization [33, 34] scale with the squareness of the loop when membranes are operated in remanence. For Co- and CoFeB-based magnetic templates (see Sec. III C), the scaling factors are 0.66 and 0.84, respectively, correspondingly reducing the current asymmetry of Eqs. (17) and (18), although the modification does not change the order of magnitude of the signal.

B. Transmitted current asymmetry

We apply this model to a beam with kinetic energy $E = 2$ eV that crosses a membrane with 7-ML graphene and 3.5-nm Co [see Figs. 6(c) and 6(d)]. For this energy beam, Ref. [17] reports an experimental Sherman function $S = 0.41$. Using $|P| = 0.25$, $T = 0.85$ [from Eq. (2) and Ref. [27]], and $N_b = 0.28$ (taken on the 7ML-MBE sample), the beam asymmetry after the membrane is $A = 0.070$.

It is worthwhile noting that the presence of broken membranes does not drastically affect the asymmetry, contrary to what could be intuitively guessed: in the ideal case with $N_b = 0$, the asymmetry would be 0.102, less than 1.5 times larger. If a larger fraction of membranes was broken, A would further decrease. For example, when doubling N_b from 0.28 to 0.56, the beam asymmetry reduces from $A = 0.070$ to $A = 0.041$, a reduction of less than half. Even in this more drastic case, where more than 50% of membranes were broken, the decrease of A would not be so critical.

When increasing the kinetic energy to $E = 6$ eV (at larger energies, the spin asymmetry of the Co electron bands almost disappears, and so S and A should do too) [35], A would reduce to 0.039 (assuming $N_b = 0.28$ and $S = 0.41$, as before), so even the energy influence is not so drastic.

The current asymmetry $\Delta I = (I_+ - I_-)$, which is the quantity to be experimentally measured, can be rewritten as

$$\Delta I = A(I_+ + I_-) = 2A\bar{I}, \quad (22)$$

where $\bar{I} = (I_+ + I_-)/2$.

Taking $E = 2$ eV and $N_b = 0.28$, so that $A = 0.070$, the current asymmetry results $\Delta I = 0.14\bar{I}$.

To be practically measurable, the standard deviation $\sigma_{\Delta I}$, due to the instrumental accuracy, must be smaller than the signal itself, i.e., $\sigma_{\Delta I} < \Delta I$. This condition defines a limit on the average beam current $\bar{I}$ that must impinge on the Faraday cup,

$$\bar{I} > \frac{\sigma_{\Delta I}}{2A}. \quad (23)$$

Assuming $\sigma_{\Delta I} > 10$ pA, we obtain $\bar{I} > 71$ pA at $E = 2$ eV and $\bar{I} > 122$ pA at $E = 6$ eV, with $N_b = 0.28$, as above. Operatively, we can estimate the signal-to-noise ratio (SNR) of the spin asymmetry signal as

$$\text{SNR} \sim \frac{\Delta I}{\sigma_{\Delta I}} = \frac{2A\bar{I}}{\sigma_{\Delta I}}. \quad (24)$$

Any transmitted current $\bar{I}$ larger than 0.75 nA at $E = 2$ eV will give a spin asymmetry signal with a signal-to-noise ratio equal to or larger than 10, thus largely acceptable.

Because of the strong attenuation of the signal below $E \sim 7$ eV due to the electron beam divergence [see Fig. 6(d)], as discussed above, at both 2 and 6 eV the measured transmitted signal $\bar{I}$ is relatively small (approximately 100 pA), and this makes the expected current spin asymmetry comparable with the experimental accuracy ($\Delta I \sim \sigma_{\Delta I}$). A current spin asymmetry $\Delta I = 6.0$ pA was effectively obtained using a beam energy of 6 eV and a transmitted current of $\bar{I} \sim 100$ pA, and the signal was acquired and stable for up to 1 h (see Appendix B for more details). Even if the signal is comparable with the standard deviation $\sigma_{\Delta I}$ of 10 pA expected from the instrumental accuracy, the preliminary result suggests the proper operation of our spin filter.

C. Electron spin polarimetry

If a larger transmitted current, above the cutoff limit of 0.75 nA, could be achieved instead, our model proves that the proposed filter would be able to act as a spin polarimeter, promising an efficiency comparable with or even larger than those reported in the literature [15–17] while relying on a relatively simple fabrication process.

Consider an electron beam with unknown polarization P_i along an arbitrary direction (i axis), i.e., coming from the exit slit of a hemispherical analyzer during an ARPES experiment and corresponding to a well-defined $(E, \mathbf{k})$ point. The electron trajectories are parallel and are incident perpendicularly on the membrane. The transmitted currents I_+ and I_- are measured using an external field (about 10 mT) to switch the membranes to have their magnetization either antiparallel (+) or parallel (−) to the i axis. Using Eqs. (20) and (22), the polarization P_i can be computed as

$$P_i = \frac{\Delta I}{2S\bar{I}} \left[1 + \frac{N_b}{(1 - N_b)T} \right] = \frac{1}{S} \frac{I_+ - I_-}{I_+ + I_-} \left[1 + \frac{N_b}{(1 - N_b)T} \right] \quad (25)$$

or, in the ideal case with all intact membranes,

$$P_i = \frac{\Delta I}{2S\bar{I}} = \frac{1}{S} \frac{I_+ - I_-}{I_+ + I_-}. \quad (26)$$

To quantify the potential of the proposed solution for polarimeters [7,36] in an ideal S-ARPES experiment, the figure of merit ε is introduced,

$$\varepsilon = \left(\frac{I}{I_0} \right) S^2. \quad (27)$$

Given the Sherman function S [17], we use Eq. (14) for the I/I_0 ratio,

$$\frac{I}{I_0} = G(1 - N_b)T(1 + P_i S) + GN_b. \quad (28)$$

In the ideal case of intact membranes ($N_b = 0$), this expression simplifies to

$$\frac{I}{I_0} = GT(1 + P_i S). \quad (29)$$

A reasonable approximation for the geometrical factor G [see Eq. (4)] in the ARPES setup is needed. Considering the region of the CCD camera corresponding to the given $(E, \mathbf{k})$ point of the spectrum, and assuming an electron beam perpendicular to the CCD, the only geometrical attenuation term is given by $G_0 = 0.25$, while G_1 can be considered unitary by design for no dispersion of the beam outside the CCD.

Assuming $S = 0.41$, $T = 0.85$ (at $E = 2$ eV), and $P_i = 0.25$, we obtain $I/I_0 \sim 0.23$ and $\varepsilon \sim 4 \times 10^{-2}$. This value is larger than those reported by competitors, being roughly 2 orders of magnitude larger than that in Ref. [15] ($\varepsilon \sim 7 \times 10^{-4}$) and about 5 to 7 times larger than in Ref. [16] ($\varepsilon \sim 8.5 \times 10^{-3}$) and Ref. [17] ($\varepsilon \sim 6 \times 10^{-3}$).

Moreover, even in the case of a beam that is not perfectly parallel and normally incident but has an angular dispersion of up to 2° , the estimated efficiency only reduces to 0.6–0.7 times the parallel one (see Appendix A for calculations), thus remaining competitive with respect to literature reports, with the additional advantage that our solution is definitely simpler to fabricate and integrate in the analyzer setup. We also observe that, for such an application, the presence of holes reported in the main text (see Fig. 3) is not a severe problem, making it possible to switch off the corresponding pixels in the CCD matrix without remarkably affecting the overall efficiency.

It is finally worthwhile highlighting the potential of the setup proposed for spin polarimetry. Because of the restricted energy window where the transmission asymmetry [see Eq. (11)] of the magnetic layer is remarkably larger than zero (around 2 eV in Co-based membranes) [35], the membrane-based spin filter is intrinsically monochromatic. To maximize the polarimeter performance, proper electron optics could be employed to accelerate/retard the beam to the energy giving the maximum value of the $\Delta I/\bar{I}$ ratio [see Eq. (26)] prior to membrane. With this tool, a full $P_i(E)$ characterization of the monochromatic electron beam could be performed, e.g., in Auger electron spectroscopy experiments where the electron energy is independent of the exciting source one.

In the case of a nonmonochromatic beam, as in the aforementioned ARPES experiments with E - $\mathbf{k}$ dispersion, we must consider that only a small portion of dispersed electrons will be efficiently filtered: as a matter of fact, to maintain the transmission asymmetry A within 15% of the peak value, an energy range of less than 1 eV of the transmitted electrons must be addressed [35]. Working at fixed photon energy, this translates to an energy dispersion of less than 1 eV [37]. If a larger energy dispersion is looked

for, tuning the photon energy could allow the beam energy to be maintained at the optimal value, so to work with a constant transmission asymmetry. Looking at k dispersion, instead, as in ARPES or reflective small-angle electron scattering experiments [38,39], the monochromaticity of the beam is intrinsically preserved so that spin resolution can be directly implemented by our system without any modification.

We also comment on the possible application of the proposed setup to the spin filtering of an unpolarized impinging beam ($P = 0$). Using Eq. (20) and the experimental data reported in Sec. IV B ($E = 2$ eV, $S = 0.41$, $T = 0.85$, $N_b = 0.28$), we obtain an asymmetry $A = 0.28$ for the transmitted beam, which increases up to 0.41 in the ideal case (100% intact membranes), in both cases more efficient than the beam polarization produced by conventional GaAs-based photocathodes illuminated by circularly polarized light [40]. To work at 2-eV kinetic energy, a proper design of the electron gun could be exploited, with the membrane grounded and sitting just between the photoelectron source, placed at -2 eV, and the axisymmetric electron optics, so that the acceleration process is made on the already polarized beam. In all the experiments using spin-polarized beams to probe the sample electronic states, such as spin-polarized inverse photoemission spectroscopy, this solution could allow us to flexibly tune the spin component probed by the beam through a proper choice of the magnetic template (without any modification of the geometry, which can be axisymmetric in all cases, at variance with conventional systems where a rotator must be introduced in the case of in-plane magnetized samples) and direction of the magnetic remanence in the membranes, allowing us to exploit a full three-dimensional reconstruction of the spin configuration of empty electronic bands.

V. CONCLUSIONS

We have demonstrated the feasibility of growing ultrathin magnetic films on suspended ultrathin graphene layers with the desired magnetic properties. We have shown that a few monolayers of graphene (7 ML) may act as a robust support for a heterostructure composed of Au(2 nm)/Co(3 nm)/Au(2 nm), leading to a survival rate of about 0.95 if the deposition is performed by magnetron sputtering, while with molecular beam epitaxy it is limited to 0.74. By contrast, if thinner graphene layers (2 ML) are used, the membranes are highly fragile (the process yields are between 50% and 75% and with a larger deviation of up to 34%). In both cases the mechanical response of the membranes is elastic up to a breaking load, which is 7.3 μ N for the 7-ML graphene thickness, and 1.3 μ N for the 2-ML one.

We have provided evidence that different deposition techniques, namely molecular beam epitaxy and magnetron sputtering, are suitable to produce such freestanding

membranes. While magnetron sputtering gives a larger process yield, molecular beam epitaxy can guarantee a subnanometric control of the roughness, down to 0.16 nm.

From the magnetic point of view, we have proved the possibility of guaranteeing a stable three-dimensional control of the magnetization reaching both in-plane and out-of-plane anisotropy with the heterostructures Au(Ru)/Co/Au(Ru) and Ta/CoFeB/MgO/Ta, respectively, which do not exceed 10 nm in thickness and have a coercive field of 3 mT for both configurations.

Electron beam transmission and filtering were finally addressed: a proper model was developed, handling transmission through intact and broken membranes, beam properties (size, divergence, spin polarization), and device geometry. We successfully compared its predictions with spin-integrated experimental data, obtaining a good qualitative fitting of the observed trend. Despite the intrinsic beam divergence produced by our setup setting a cut-off for the transmission at about 7 eV, we were able to provide a preliminary indication of spin filtering. We conclude that spin polarimetry is within reach, with an efficiency comparable to or even exceeding that of existing devices, while maintaining a relatively simple fabrication process.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request [41].

APPENDIX A: POLARIMETER PERFORMANCE FOR DIVERGENT BEAMS

Consider the side view of the heterostructure sketched in Fig. 7 (additionally, see Fig. 1 in the main paper). In this section, the beam propagation across the Cu support grid is modeled to provide insights about the figure of merit of the filter, considering a cylindrical symmetry for both the membrane and the beam and considering the angular divergence of the beam. Given the diameter D and thickness H of the cylindrical hole in the copper support layer

underneath a membrane, an electron crossing the membrane template (graphene + buffer, magnetic and capping layers) could exit from the bottom side without colliding with the Cu walls if its incidence angle is below a certain value.

Consider an electron beam arriving at the entrance hole at a fixed incidence angle θ [Fig. 7(b)]. Only electrons within the segment AB will arrive at the exit without colliding against the side walls. The fraction of transmitted electrons can be calculated as the ratio between the AB length and the hole diameter,

$$f = \frac{\overline{AB}}{\overline{AC}} = \frac{d}{D} = \frac{D - H \tan \theta}{D} = 1 - \frac{H}{D} \tan \theta. \quad (\text{A1})$$

This fraction depends on θ and is unitary, as expected, for a perfectly parallel beam with a null incident angle (all the electrons pass through the hole), while for any $\theta \geq \theta_{\max}$ it is zero (nothing can pass through). $\theta_{\max}$ is the maximum angular aperture that allows electrons to not collide with the walls and can be calculated referring to the situation illustrated in Fig. 7(a),

$$\theta_{\max} = \arctan\left(\frac{D}{H}\right) \sim 3.4^\circ. \quad (\text{A2})$$

We can rewrite Eq. (A1) more formally as follows:

$$f(\theta) = \begin{cases} 1 - \frac{H}{D} \tan \theta & \forall \theta < \theta_{\max} \\ 0 & \forall \theta \geq \theta_{\max} \end{cases}. \quad (\text{A3})$$

In the main text, we demonstrated that the beam intensity after crossing the membrane setup, assuming normal

incidence of the beam, is given by [see Eq. (29)]

$$I = I_0 GT(1 + P_i S), \quad (\text{A4})$$

where I_0 is the total beam intensity before the membrane setup, G is the geometrical attenuation factor, S is the Sherman function, and P_i is the beam polarization along the i axis. We assume all the membranes are intact.

To introduce the θ dependence in the transmitted current, Eq. (A4) is multiplied by the factor $f(\theta)$ of Eq. (A3) to obtain the intensity collected by the detector when a parallel beam impinges at an angle θ ,

$$I(\theta) = I_0 GT(1 + P_i S) f(\theta). \quad (\text{A5})$$

The effect of a beam with a variable angular dispersion of electrons is then introduced using the probability density function $p(\theta)$, so that $p(\theta) d\theta$ is the probability that an electron of the beam hits the membrane with an incidence angle between θ and $\theta + d\theta$.

The total transmitted intensity is thus obtained by integrating the product $I(\theta)p(\theta)$ on the angular range $[0, \theta_{\max}]$,

$$\begin{aligned} I &= \int_0^{\theta_{\max}} I(\theta) p(\theta) d\theta \\ &= I_0 GT(1 + P_i S) \int_0^{\theta_{\max}} f(\theta) p(\theta) d\theta. \end{aligned} \quad (\text{A6})$$

Using Eq. (A3), we obtain

$$\begin{aligned} I &= I_0 GT(1 + P_i S) \int_0^{\theta_{\max}} \left(1 - \frac{H}{D} \tan\right) p(\theta) d\theta \\ &= I_0 GT(1 + P_i S) F. \end{aligned} \quad (\text{A7})$$

In the last step, we defined the function F as

$$F = \int_0^{\theta_{\max}} \left(1 - \frac{H}{D} \tan\right) p(\theta) d\theta, \quad (\text{A8})$$

which determines the final value of I given the values of the other parameters (I_0, G, T, P_i). In the following, two different probability density functions $p(\theta)$ are considered and their results are compared.

1. Constant probability density

We assume that all angles between 0 and θ_0 are equally probable,

$$p(\theta) = \begin{cases} p & \forall \theta \leq \theta_0 \\ 0 & \forall \theta > \theta_0 \end{cases}, \quad (\text{A9})$$

with $\theta_0 < \theta_{\max}$. This assumption corresponds to considering a beam with a maximum incidence angle θ_0 .

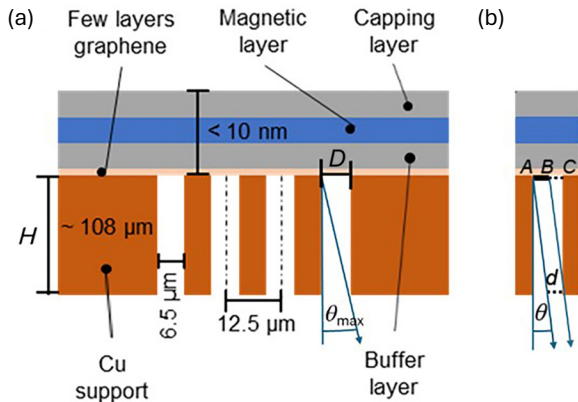


FIG. 7. (a) Cross-sectional view of the heterostructure on top of the Cu support. (b) Sketch of the single membrane crossed by electrons impinging at an incident angle θ with respect to the normal.

The integral can be analytically solved as follows:

$$\begin{aligned} F &= \int_0^{\theta_0} \left(1 - \frac{H}{D} \tan\theta\right) p \, d\theta \\ &= p \left(\theta_0 + \frac{H}{D} \ln|\cos\theta_0|\right). \end{aligned} \quad (\text{A10})$$

The value of p can be found by imposing the normalization condition,

$$\int_0^{+\infty} p(\theta) d\theta = 1. \quad (\text{A11})$$

Exploiting the definition of $p(\theta)$ in Eq. (A9) we achieve

$$\int_0^{+\infty} p(\theta) d\theta = \int_0^{\theta_0} p(\theta) d\theta = p \int_0^{\theta_0} d\theta = 1, \quad (\text{A12})$$

so that

$$p = \frac{1}{\theta_0}. \quad (\text{A13})$$

We finally obtain

$$F = 1 + \frac{H}{D} \frac{\ln|\cos\theta_0|}{\theta_0}, \quad (\text{A14})$$

which represents the fraction of the incidence beam that overcomes the membrane setup and arrives at the detector. Finally, the total intensity [Eq. (A7)], the I/I_0 ratio, and the figure of merit ε of the polarimeter [Eq. (27) of the main paper] are, respectively,

$$I = I_0 GT(1 + P_i S) \left(1 + \frac{H}{D} \frac{\ln|\cos\theta_0|}{\theta_0}\right), \quad (\text{A15})$$

$$\frac{I}{I_0} = GT(1 + P_i S) \left(1 + \frac{H}{D} \frac{\ln|\cos\theta_0|}{\theta_0}\right), \quad (\text{A16})$$

$$\varepsilon = \left(\frac{I}{I_0}\right) S^2 = GT(1 + P_i S) S^2 \left(1 + \frac{H}{D} \frac{\ln|\cos\theta_0|}{\theta_0}\right). \quad (\text{A17})$$

Note that all these results are scaled by the factor F with respect to the normal incidence ones (corresponding to setting $\theta_0 = 0^\circ$). To give a quantitative estimation of F , we assume $\theta_0 = 2^\circ$, a quite realistic and conservative value for common electron analyzers. As in the main paper, we use $E = 2$ eV, $S = 0.41$, $T = 0.85$, and $P_i = 0.25$. The results are reported in Table II. The final efficiency is only reduced by a factor of around 0.7 with respect to a perfectly parallel normally incident beam, leaving unchanged the supremacy of our proposed solution with respect to competitors [15–17].

TABLE II. Main parameters and related efficiency for different beam configurations.

	Normal incidence	Constant $p(\theta)$	Gaussian $p(\theta)$
E (eV)	2	2	2
S	0.41	0.41	0.41
T	0.85	0.85	0.85
P_i	0.25	0.25	0.25
θ_0 (deg)	0	2	2
$F(\theta_0)$	1	0.71	0.56
I/I_0	0.23	0.17	0.13
ε	3.9×10^{-2}	2.8×10^{-2}	2.2×10^{-2}

2. Gaussian probability density

We assume that the incidence angle of an electron follows a Gaussian functional distribution,

$$N(\theta) = \frac{1}{\sqrt{2\pi}\sigma} e^{-\frac{\theta^2}{2\sigma^2}}, \quad (\text{A18})$$

where we take $\sigma = \theta_0$, so that around 69% of electrons impinge on the membrane with $\theta \leq \theta_0$. With respect to the previous model, the electron incidence angle can be larger than θ_0 , but with lower probability. Using this distribution, we account for the evidence that most of the electron paths are almost parallel and normally incident, and only a few are divergent.

The probability density function $p(\theta)$ can be derived from the normalization condition [Eq. (A11)] considering that:

(1) $p(\theta)$ must be integrated from 0 to $+\infty$, so that, using the normalization condition on $N(\theta)$,

$$\int_{-\infty}^{+\infty} N(\theta) d\theta = 2 \int_0^{+\infty} N(\theta) d\theta = \int_0^{+\infty} p(\theta) d\theta, \quad (\text{A19})$$

we obtain

$$p(\theta) = 2N(\theta). \quad (\text{A20})$$

(2) Integration over θ larger than $\theta_{\max}$ has no impact because $f(\theta) = 0$ [see Eq. (A3)], so that, as the upper integration bound, we can take $\theta_{\max}$ instead of $+\infty$.

The integral for F finally gives

$$\begin{aligned} F &= \int_0^{\max} \left(1 - \frac{H}{D} \tan\theta\right) p(\theta) d\theta \\ &= \int_0^{\max} \left(1 - \frac{H}{D} \tan\theta\right) \frac{2}{\sqrt{2\pi}\sigma} e^{-\frac{\theta^2}{2\sigma^2}} d\theta. \end{aligned} \quad (\text{A21})$$

The numerical solution is reported in Fig. 8 (black full dots) for discrete values of θ_0 in the $[0, \theta_{\max}]$ range.

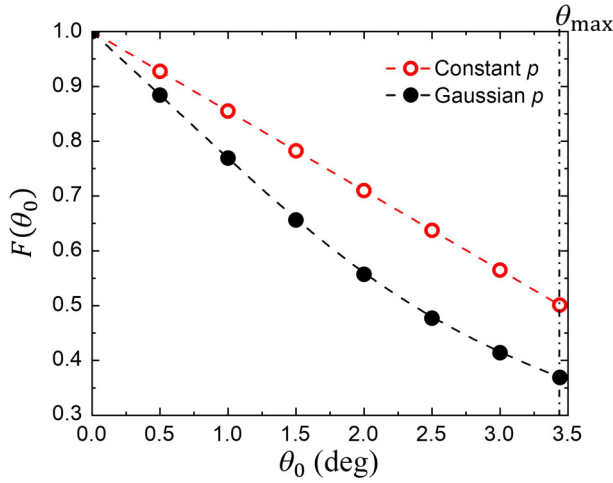


FIG. 8. Fraction F of the incidence beam that overcomes the membrane setup and arrives at the detector, calculated assuming constant (red circles) or Gaussian-like (black dots) probability densities.

For comparison, the corresponding values obtained with the constant probability density (Sec. A1) are also indicated (red circles). Note that the final efficiency with the Gaussian distribution is smaller than with the constant one because of spreading to larger angles than θ_0 . Taking $\theta_0 = 2^\circ$, as reported in Table II, the efficiency is globally reduced by a factor of around 0.6 with respect to a perfectly parallel normally incident beam.

Before we conclude, we comment that a realistic result should require a precise definition of the beam profile, which is outside the scope of this paper. The result obtained with the Gaussian probability distribution can anyway be considered a smaller edge for the efficiency, because it is calculated in a quite extreme configuration where the beam spreads well over the θ_0 angle. If, more likely, we would take $2\sigma = \theta_0$ ($3\sigma = \theta_0$), so that more than 95% (99.5%) of electrons would impinge on the membrane with $\theta \leq \theta_0$, the efficiency would increase to 0.77 (0.85), compared with 0.56 when $\sigma = \theta_0$. This trend suggests that the electron beam spreading is not so detrimental to spin detection, confirming that our proposed technology deserves real potentiality for S-ARPES experiments with real-time E - $\mathbf{k}$ - $\mathbf{s}$ resolution.

APPENDIX B: SPIN ASYMMETRY MEASUREMENT

Figure 9 shows the spin symmetry ΔI measured on the membrane with 7-ML graphene and 3.5-nm Co, using a beam energy of 6 eV and a transmitted current $\bar{I} \sim 100$ pA. Each measurement runs for about 1 h to mimic typical ARPES acquisition times, during which the current spin asymmetry ΔI is acquired at a frequency of 3 Hz. The spin asymmetry of the transmitted beam is comparable in each

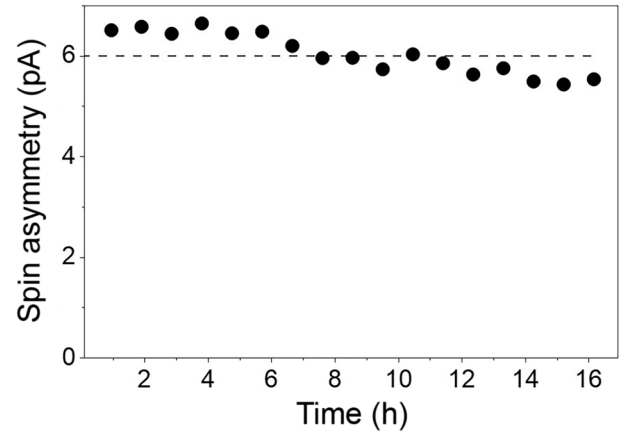


FIG. 9. Spin asymmetry of the transmitted beam (dots) and time averaged value (dashed line).

run, and only a relatively small decrease (approximately 1 pA) is observed over 16 h, possibly due to mechanical, optical, or electrical drifts over such a long measurement time.

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