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Rashba-like spin textures in Graphene promoted by ferromagnet-mediated Electronic-Hybridization with heavy metal

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Epitaxial Graphene/Ferromagnetic (Gr/FM) structures deposited onto heavy metals (HM) have been proposed for the realization of novel spin-orbitronic devices because of their perpendicular magnetic anisotropy and sizeable Dzyaloshinskii-Moriya interaction, which in turn allow for enhanced thermal stability and stabilization of chiral spin textures. In this work we elucidate the nature of the induced Spin-Orbit Coupling (SOC) at Gr/Co interface on Ir by investigating different FM thicknesses. Angular and Spin-Resolved Photoemission Spectroscopy and Density Functional Theory experiments show that the interaction of the HM with the C atomic layer via hybridization with the FM is indeed the source of the SOC in the Gr layer. Furthermore, our studies in ultra-thin (2 ML) Co film underneath Gr reveal an energy splitting of ~100 meV (negligible) for *in-plane (outof-plane)* spin polarized Gr π bands, consistent with a Rashba-SOC at the Gr/Co interface, which is either the fingerprint or the origin of the Dzyaloshinskii Moriya interaction. Interestingly, at larger Co thicknesses (~ 10 ML), neither *in-plane* or *out-of-plane* spin splitting is observed, indicating Gr is almost decoupled from the HM.

INTRODUCTION

Spintronics aims at exploiting the spin degree of freedom of electrons for new forms of information storage and logic devices [1]. A major challenge for innovative, high-speed, low-power operation spintronic-based devices is to develop suitable spin transport channels with long spin lifetime and propagation length. Graphene (Gr) is an optimal spin channel material, exhibiting the longest spin-relaxation length ever measured at room temperature (RT) of several micrometers [2-7], and enhanced spin to charge conversion and spin Hall effect when interfaced with heavier atoms [8, 9]. This is due to the Spin–Orbit Coupling (SOC), which is an essential spin interaction that relates the spin to the momentum of electrons. While, from one side, this interaction can drive to a modification of spin relaxation or spin diffusion length, strong SOC also dictates a vast variety of related intriguing phenomena, especially when reduced dimensions are at place. Few examples are quantum spin Hall and anomalous Hall states [10–16] or skyrmions [17, 18].

However, Gr is a material that presents a negligible SOC in its pristine state. At the K point of the Gr Brillouin Zone (BZ), where the Dirac point is located at the Fermi level (E_F), the spin-splitting is very small, of only 24-50 μ eV [19]. To observe spin-orbit effects suitable for applications it is necessary to induce SOC in Gr while not destroying its Dirac cone structure. By proximity, i.e., by combining Gr with the appropriate substrates [20–23], or by intercalation of heavy atoms in suitable geometries [24–28], a strong hybridization and spin-splitting near the Dirac point can be deliberately induced. For instance, gold intercalation at the Gr/Ni interface has been demonstrated to create a giant spin–orbit splitting (~ 100 meV) of the Gr Dirac cone up to the Fermi energy [24], attributed to a Rashba effect (*in-plane* spin polarization). Besides, in the case of Gr/Pb grown onto Pt(111) [26, 29], the observed enhancement of SOC has been explained in terms of intrinsic (Kane and Mele type [11]) and extrinsic (Rashba [25]) SOC contributions, responsible for *out-of-plane* and *in-plane* spin-polarizations, respectively.

The intercalation of ferromagnetic (FM) thin layers in Gr enables, furthermore, the tuning of the spin filtering and injection efficiency [30–32], exchange coupling [33], tunnel magnetoresistance [34], Rashba effect [25, 35, 36] and perpendicular magnetic anisotropy (PMA) [37–42], as well as the stabilization of magnetic skyrmions [43, 44]. However the hybridization with the metal states is so strong that the Dirac cone results destroyed at the Fermi level [45–48].

Recently, we have engineered novel high quality Grbased PMA heterostructures with atomically flat interfaces and a homogeneous epitaxial Co-intercalated layer, sandwiched between epitaxial Pt(111) or Ir(111) buffers and a Gr monolayer [38, 41, 49]. In these systems, the hybridization of Co 3d orbitals with Gr bands enhances PMA [38, 39] via the anisotropy of the orbital magnetic moments [38] and, rather unexpectedly, originates a large chiral Dzyaloshinskii-Moriya Interaction (DMI), even at the Gr/Co interface. In contrast to Co/Pt interface, in which the DMI is explained by a Fert-Levy model [50, 51], the physical origin of the DMI at Gr/Co has been attributed to a Rashba mechanism [40, 41]. It is worth mentioning that very recent DFT calculations revealed that the DMI at both Gr/Co and Co/Pt interfaces may have the same nature, where the effect of Gr is to reduce the DMI of the heterostructure by the inversion of the chirality of the vacuum/Co interfacial DMI [52].

Here we report on the study of the electronic structure of epitaxial Co-intercalated Gr layers on Ir(111)grown onto insulating oxide substrates [49], as a function of the Co thickness, with unprecedent resolution of the electronic states involved at the interface close to the Fermi level. In order to unravel the electronic origin of the SOC induced in the Gr π band structure near the Dirac point in epitaxial Gr/Co/Ir multilaver, we have performed ARPES and SR-ARPES experiments combined with Density Functional Theory (DFT) calculations. We have found that: i) independently from the Co thickness, up to ~ 10 monolayers (ML), the electronic structure of Gr reflects a highly epitaxial Co intercalation and a quasi-commensurate lattice with respect to Gr/Ir system; ii) at low Co thickness (i.e., 2 ML), the pure interfacial Co 3d states hybridize at both sides of the interface, i.e., with Ir 5d and Gr π bands and, hereby, the interaction of the HM layer with Gr via the intercalated FM atoms can be assigned as source of SOC for C atoms; iii) SR-ARPES measurements and DFT calculations reveal an *in-plane* energy spin-splitting of $\sim 100 \text{ meV}$ for the thin Co (2 ML), whereas it is lost at larger Co thicknesses (~ 10 ML), in agreement with the prediction for three or more intercalated Co layers [52].

RESULTS AND DISCUSSION

Electronic structure of $Gr/Ir(111)/Al_2O_3(0001)$

The procedure adopted for the preparation of epitaxial Gr-based heterostructures is reported in reference [49] and in Methods section. The samples were entirely grown and meaured in UHV conditions. In brief, 30 nm thick Ir buffers were DC sputtered at 670 K on Al₂O₃(0001) single crystals, onto which the Gr epitaxial monolayer was subsequently grown by ethylene dissociation at 1025 K. The Gr/Ir(111) structure was demonstrated to exhibit the well known ~ 10 × 10 Moiré pattern due to the coexistence of the incommensurate unit cells of Ir and Gr [38, 49, 53] (see LEED patterns in Figure 2(a)).

The electronic structure of Gr/Ir(111) along $\overline{\Gamma KM}$ direction is depicted in Figure 1 (a,b,c) and it is consistent with the expected quasi-free standing character of the Gr layer on Ir(111), and comparable to the case of Gr grown on Ir(111) monocrystals [28, 54–56], proving the high quality of the interfaces. The measured Gr σ and π bands are very sharp and intense, as evidenced by the Momentum Distribution Curve (MDC) at -150 meV, and the Energy Distribution Curve (EDC) profiles at the Kpoint of Gr BZ (panels (d) and (e), respectively), confirming again the interface quality. The dominant feature in the spectrum is the Gr π band. The bottom of the π band at Γ is located at a Binding Energy (BE) of -8.28 eV and disperses towards E_F , resulting in the formation of a slightly n-doped Dirac cone (panel (a) and zoom in (b)) as inferred from EDC and MDC analyses (panels (d) and (e)). The narrowest MDC corresponds to a BE of -150 meV and can be fitted with a lorentzian function (panel (d)). The full width at half maximum (FWHM) is $\sim 0.0061 \text{ Å}^{-1}$, similar to the FWHM value obtained for Gr grown on Ir(111) single crystals [55]. Moiré π band replicas are not clearly well resolved because of the strong intensity of the main Gr π band at this photon energy $(h\nu=64 \text{ eV})$ and the fact that the Moiré pattern is not fully developed (see LEED pattern in Figure 2 (a)). The Dirac cones are also visible at the Fermi Surface (FS) and Constant Energy (CE) maps in Figure 1 (f).

Here, the Dirac cones at K points, indicated by red dashed lines, show the characteristic trigonal symmetry of quasi-free standing Gr grown on Ir(111) and coexist with the expected Ir(111) single crystal electronic structure, as earlier reported [47, 57]. Note that these main Ir 5d ellipsoidal-shaped features close to E_F are also evidenced in the curvature ARPES band maps (panel (a) of figure 2). The Gr BZ and Ir Surface BZ (SBZ), indicated by red and orange hexagons, are clearly visible in figure 1 (f). Additionally, hybridization of Ir 5d bands with Gr π band is evidenced in the BE region between -1 and -4 eV, resulting in some discontinuities in the dispersion along $\overline{\Gamma K}$ direction. Nevertheless the hybridization effects are not strong enough in such a way that the Dirac-like dispersion and the Dirac cone are preserved.

Electronic structure of Co intercalated on Gr/Ir

Two Co layer thicknesses (i.e., 2 and 10 ML) have been investigated. Co was grown by e-beam deposition evaporated at RT and low deposition rate, while the intercalation in Gr was promoted by a gentle thermal annealing (see Methods section). The completion of the intercalation process was checked by monitoring the LEED pattern, since this is lost when Co islands are on top of Gr and then recovered after Co completely intercalated at the Gr/Ir interface [41, 49].

Figure 2 (and S1) shows the electronic band structure



FIG. 1. Electronic structure of Gr/Ir(111). (a) ARPES intensity band map along Γ KM direction. (b) Zoom on the Dirac cone corresponding to the red rectangular area in panel (a). (c) Perpendicular cut to Γ K direction centered at K point. (d) MDC and (e) EDC profiles at red dashed lines indicated in panels (b) and (c). (f) FS and CE maps. Red dashed lines indicate the Gr Dirac cones. Ir SBZ and Gr BZ are indicated by orange and red hexagons, respectively.

along $\overline{\Gamma M K \Gamma}$ high symmetry directions of the Gr single layer on Ir(111) and Co intercalated on Gr/Ir.

The LEED patterns in the side panels of figure 2 (a) and (b) exhibit signatures of a Moiré pattern and a clear Gr structure is observed in panel (c), indicating in all cases the Co complete intercalation. As mentioned above, the Gr/Ir(111) electronic structure shows the typical band dispersion of quasi-free standing graphene for π and σ bands. At this photon energy, the dispersing bands originating from Ir 5d states in the E_F to -4 eV energy range consist mainly of the Rashba split surface state dispersing towards Γ [59] and the two concentrical ellipsoidal pockets centered at M_{Ir} point of Ir Surface BZ (SBZ) [47, 57], clearly visible in the FS of Gr/Ir, Figures 1(f) and 4 (a).

Upon Co intercalation, we notice that the electronic band structure of Gr becomes highly modified because of the strong interaction with the underlying Co layer. For the 2 ML Co, the π band is negatively energy shifted by ~ 2 eV (see Figure 2 (b), and Figures S1 and S2 in Supplementary Information). At Γ point, the bottom of the π band is shifted from -8.28 to -10.08 eV and merges with the Co 3d states at ~ -2.80 eV, close to K point. The intensity and sharpness of the Gr bands are reduced with respect to the Gr/Ir, but the π band dispersion (also σ) can be still easily followed.

Interestingly, further Co intercalation results in a double-like π band dispersion as appreciated in the maps in Figure 2 (c), and in the EDC profiles extracted at the green dashed lines in Figure 2 and reported in Figure S2. The double π band can be interpreted as a non-perturbed Gr/Ir π -band closer to E_F plus the more electronically doped π' Gr/Co at deeper BE (associated with a Cointercalated Gr/Ir scenario). As reported earlier [38], in fact, although the Co intercalation underneath Gr is layer-by-layer and epitaxial (as confirmed here by the measured electronic structure, Figures 2, S1, 3 and 4), the formation of Co clusters or inhomogeneities at large thicknesses cannot be completely ruled out and can eventually lead to a more corrugated Gr layer. The doublelike π band component can thus be associated to a corrugated Gr layer where the π feature would appear due



FIG. 2. Curvature ARPES band maps: (a) electronic structure of Gr/Ir(111), (b) intercalated Gr/2 ML Co/Ir and (c) intercalated Gr/10 ML Co/Ir(111) along $\overline{\Gamma M K \Gamma}$ high symmetry directions. Red and orange dashed lines follow the π and σ bands for Gr/Ir(111) and Gr/2 ML Co/Ir(111), respectively. On the left, the schematic sketches of the corresponding heterostructures and their LEED patterns obtained at energies of 69 eV (Gr/Ir(111) and Gr/2 ML Co/Ir) and 155 eV (10 ML Co). Note that the ARPES band maps were obtained after applying the curvature algorithm [58] in order to obtain a sharper and better resolved electronic band dispersion (raw data are shown in Supplementary Information, figure S1). Green dashed lines in the band maps indicate the position at which the EDC profiles have been extracted (Figure S2).

to weakly interacting, hill, C atoms, while the π' band would arise from highly interacting, valley, C atoms.

As indicated by the EDC profiles in Figure S2, this doublet dispersing state matches with the sum of highly interacting Gr-Co π' states and less interacting Gr-Ir π band (blue and red dashed lines), as also reported for similar systems [46].

Besides, in Figures 2 (b) and (c) several prominent Co electronic states can be identified: *i*) a narrow and highly intense Co peak close to E_F along $\overline{\Gamma M K \Gamma}$ in a BE range between -0.2 and -0.5 eV, common to the two Co thicknesses considered; *ii*) for the thicker Co system (figure 2 (c)), a highly localized and broad level located at -8.3 eV is present in the band structure, which could be indicative of a more disordered thicker, although epitaxial, Co layer; *iii*) another strongly dispersing band from -1.9 eV to E_F at Γ , which confirms that, apart from the possible disorder, the intercalation of the thickest cobalt layer is mainly epitaxial; *iv*) other localized states can be detected for the 10 ML Co at -1.1 and -2.9 eV; *v*) the Ir features disappear at larger Co thickness.

In the following we provide a closer inspection into the



FIG. 3. Zoom on the bands close to E_F for (a) Gr/Ir(111) and (b) Gr/2 ML Co/Ir(111) along $\overline{\Gamma M}$ direction. Raw and curvatureprocessed ARPES band maps with and without superimposed guidelines are presented, respectively. Blue dashed lines indicate the Ir(111) bands (a), which persist and hybridize with Co states (b) as evidenced by the corresponding gap openings (a zoom of the gap region with guidelines is presented in the inset in bottom row first panel). Red and green dashed lines follow the Gr bands and a Gr replica from the Moiré reconstruction, respectively. Red arrows indicate the BEs corresponding to the CE maps shown in Figure 4.

electronic properties of the 2 ML Co system. Previous ARPES results have shown how after intercalation of one or more cobalt layers, the iridium bands close to E_F can barely be distinguished, while the Co 3d states are so intense and broad that the Ir 5*d* bands become blurred and diffuse [46, 47]. A zoom on the electronic structure from E_F to -5.5 eV for both Gr/Ir and Gr/Co (2 ML)/Ir systems along $\overline{\Gamma M}$ and $\overline{\Gamma K}$ directions is shown in Figures 3 and S3, respectively. Raw ARPES and curvature ARPES band maps are displayed for both systems. Similarly, the FS and CE maps are also presented in Figures 4 (a) and 4(b).

First, it should be noted that different bands corresponding to the electronic states of the underlying Ir(111) layer can always be detected, contrary to what previous studies have reported [46, 47]. Noticeably, the electronic states that make up the ellipsoidal electron pockets centered at M_{Ir} are unambiguously resolved at the FS (figure 4 (a)), mainly the prominent band dispersing from -3.8 eV at 0.6 Å⁻¹ to E_F at 1.13 Å⁻¹ along $\overline{\Gamma M}$ (figure 3 (b)), which coincides with the same band for Gr/Ir(111) (figure 3 (a)). Particularly, the hybridization between Co 3d states at ~ -0.5 and -1.2 eV (clearly resolved between 0.5 and 0.8 Å⁻¹ along $\overline{\Gamma M}$) and the Ir dispersing band becomes evident, as reflected in the increased intensity (red arrows on raw ARPES band maps in figure 3 (b), left) and evidenced by the gap opening observed in both, raw and curvature ARPES band maps (blue dashed lines figure 3 (b), right). The main gap opening of Ir dispersing band occurs between -1.1 and -0.65 eV at 0.99 Å⁻¹ along $\overline{\Gamma M}$ (magnified in the inset of figure 3 (b)).

This hybridization is also reflected in the CE maps at -0.5 and -1.0 eV in Figure 4 (a) and (b): the Ir ellipsoidal pockets close to E_F in Gr/Ir (panel (a)), become closed ellipses when interacting with these Co 3d states (panel (b)). It is worth noting that, besides the Co-Ir hybridization close to M_{Ir} , the Ir Rashba surface state [59], with maximum intensity at -0.26 eV at Γ , is still preserved after the Co intercalation (albeit slightly modified by the interaction with Co 3d states), as it can be gathered from both $\overline{\Gamma M}$ (Figure 3) and $\overline{\Gamma K}$ (figure S3) directions bandmaps. Thus, the Ir Rashba surface state at Γ and Ir 5d bands close to M are preserved and hybridized after the intercalation of 2 ML of Co.

Finally, one more key aspect about the electronic structure of the 2 ML thick Co system, is the development of mini Dirac cones at a narrow energy region of 0.2 eV below E_F at Gr K point (panel (c) in Figure 4). These mini Dirac cones have been previously observed in perfectly oriented Gr monolayer on a Co (0001) surface and they



FIG. 4. (a) FS and CE maps at -0.5 and -1.0 eV for Gr/Ir(111). (b) FS and CE maps at -0.5 and -1.0 eV for Gr/2 ML Co/Ir(111). Orange and red lines indicate the BZ for Ir(111) and Gr monolayer, respectively. (c) Mini Dirac cones. The profiles were taken along the perpendicular white dashed lines crossing at K point as indicated over the FS at panel (b).

strongly depend on the epitaxial quality of the interface [45]. The mini Dirac cones are due to the interaction of C $2p_z$ and Co 3d orbitals, have a fully two-dimensional character with their wave functions located at the interface and are spin polarized. The development of the minicone states is a hallmark of our interface quality and highlights both their structural and electronic properties.

In summary, after 2 ML of intercalated Co, the surface state and bulk Ir features are preserved at E_F and become strongly modified just below it because of the interaction with Co 3d electronic states. Moreover, the perfectly oriented epitaxial structure layer leads to the formation of the mini Dirac cone as a result of Co-C interaction. Further Co intercalation decouples Ir(111)electronic structure from Gr/Co interface, while some disorder effects can be introduced. From our ARPES results we have identified the Co-Ir electronic hybridization, including the Ir(111) Rashba surface state, as well as Co-C electronic interaction. Thus, we can assign the Ir(111) layer interaction with C atoms via the intercalated Co layer (at least for the lower Co thickness) as the SOC source at the Gr/Co interface. This may give rise to a DMI at Gr/Co interface, as previously suggested [52].

SR-ARPES on 2 ML Co intercalated on Gr/Ir

In order to investigate whether an extrinsic or intrinsic spin-orbit induced spin-splitting is generated in Gr by Co intercalation, we have resorted to SR-ARPES experiments and DFT calculations. SR-ARPES spectra were acquired at selected wave vector values, k, of the Gr BZ, with the aim of seeking for any sign of an energy spinsplitting occurring either for *in-plane* or *out-of-plane* spin polarization. The Spin Resolved EDCs (SR-EDCs) for the *in-plane* and *out-of-plane* components are presented in Figures 5 (b) and (c), respectively, measured at 1.48 Å⁻¹ along $\overline{\Gamma K}$, as indicated by the green dashed line in the ARPES band map (panel (a)), close to the apex of the doped main Dirac cone, and thus to obtain a considerable signal suitable for spin resolution.

To isolate and extract the precise contribution of the Gr π band, the SR-EDCs have been normalized and, after that, fitted with five components, which are (see Figure 5): the two peaks closer to E_F arising from Co 3d states (turquoise and dark blue curves in panels (b,c), and dashed lines in panel (a)); the Gr σ band contribution (purple curve and dashed line); the wide feature appearing after Co intercalation (grey, contribution from disorder or Co localized states after intercalation); and the more intense Gr π contribution (orange). This analysis has been applied to both *in-plane* and *out-of-plane*



FIG. 5. (a) ARPES band map along $\overline{\Gamma K}$. Green dashed line indicates the polar angle θ (i.e. wavevector) at which the SR-ARPES measurements on panels (b) and (c) were taken. Horizontal dashed lines indicate the BE of the fitted components of SR-EDCs curves. Normalized experimental SR-EDCs for (b) *in-plane* channels and (c) *out-of-plane* channels. Solid red and blue lines correspond to the fits of the experimental curves, while their individual components are displayed at the bottom of each graph. See the main text for more details of the origin of each component.



FIG. 6. (a) In-plane and (b) out-of-plane SR-EDCs zoon on Gr π band components for left and right (in-plane), and up and down (out-of-plane) channels, respectively. Dark red and blue solid lines correspond to the fitting of experimental result (light red and blue solid lines). Insets show the spin polarization calculated from equation 1.

components, measured simultaneously by the Spin mini-Mott detector. Further details on spin resolved measurements, data analysis and curve fitting of SR-EDCs are reported in Experimental Methods section and Supplementary Information.

As clearly seen in panels (b) and (c) of figure 5, we found an energy splitting for *in-plane* left and right spins of the Gr π bands of $\Delta E_{in-plane} = (100 \pm 40)$ meV. Con-

trarily, the *out-of-plane* spin up and down components do not show any sizeable splitting above the average error of the curve fitting procedure, $\Delta E_{out-plane} = (20 \pm 40)$ meV. A second set of SR-ARPES measurements at a close θ angle result in a similar energy splitting for *inplane* and *out-of-plane* components (see Supplementary Information).

In figure 6 the results of the fittings of Gr π spin components (zoom on the experimental data in panels (c) and (d)) for both *in-plane* (panel (a)) and *out-of-plane* (panel (b)) channels are presented separately. The corresponding spin polarization is calculated from equation 1:

$$P_{in(out)} = \frac{1}{S_{eff}} \frac{I_{left(down)} - I_{right(up)}}{I_{left(down)} + I_{right(up)}};$$
 (1)

where S_{eff} is the Sherman function, estimated to be 0.16 for this spin Mott detector, and the intensities (left, right, up, down) are the ones calculated from the fitting procedure in order to minimize the effect of the noise level of raw data. The obtained spin polarizations for both components are displayed in the insets in figure 6. The energy spin-splitting and both *in-plane* and *out-of-plane* spin polarizations are consistent with previous results on HM intercalated systems in Gr, where a Gr π band energy splitting of $\Delta E_{SOC} \sim 70\text{-}100 \text{ meV}$ was reported for Au and Pb intercalated layers at a similar k value along $\overline{\Gamma K}$ [24, 25].

Finally, SR-ARPES measurements were also performed on the thicker, 10 ML Co sample (see figure S15). The intrinsic spectral broadening of Gr π band induced by Co intercalation makes it difficult to resolve any possible spin-splitting, even if the fittings of Gr π band indicate that any possible energy spin-splitting is well below the fitting errors for both *in-plane* and *out-of-plane* components (see Supplementary Information). Such absence of measurable spin-splitting in thicker Co film highlights the essential role of the HM support, and corroborate the ARPES results that at larger Co thickness the top Gr/Co interface is almost decoupled from the bottom Co/Ir one. The *in-plane* spin-splitting observed in thinner Co may be either considered as fingerprint of, or originate, the non-zero DMI found at the Gr/Co interface, and that cannot be attributed only to individual Co-C hybrid bands near the Fermi level, but also the strong SOC of the HM must be taken into account [52].

DFT calculations on 2 ML Co intercalated on Gr/Ir

DFT calculations with spin-orbit interaction have been performed for 2 ML of epitaxial Co intercatalated in Gr $1 \times 1/\text{Ir}(111)$ interface, and confirm the experimental results. Gr 1×1 has been expanded to match with the Ir lattice, so K and M symmetry points are referred to Ir(111) SBZ. In this way, contributions from Gr Moiré superlattice to the electronic structure are explicitly neglected. The surface projected band structure of Gr/Co/Ir(111) is shown in Figure 7 where we highlight the Density of States (DOS) of Gr and Ir at the interface (panel (a)) and plot the S_y spin resolved band structure (panel (b)). The area of the circles is proportional to DOS(Gr) × DOS(Ir), being thus indicative of the hybridization between graphene and Ir. More details of the spin- and surface projected band structures are shown in the Supplementary Information (Figures S16 and S17).

After a first comparison of the DFT results with the experimental ARPES data (Figures 2 (b) and S1 (b)), several aspects can be highlighted. The Gr π bands are preserved and hybridized with Ir-Co states at the interface (rectangle in Figure 7 (a)). The main Dirac point of the Gr π band is located at ~ -3 eV, close to K, as also observed in the ARPES results (see Figures 2 (b), S1 (b) or S4 (b)). Additionally, signs of the Gr-Ir hybridization can be observed at ~ -1.5 eV and, mainly, close to E_F at K (green arrows), where the strong C-Co hybridization gives rise to the spin-polarized mini Dirac cone (as observed in figure 4 (c)). Note that the calculations neglect the band shift due to electron doping, therefore the mini Dirac cone appears slightly above the Fermi level. Besides, there is a small enhancement of DOS close to M at ~ -1.5 eV (black arrow), that is compatible with the Ir-Co hybridization detected in ARPES results (Figure 3, Figure 4 and S3).

The calculated *in-plane* spin contribution along $\overline{\Gamma K}$ is represented in Figure 7 (b). The energy and momentum ranges correspond to the rectangle in figure 7 (a). The red and blue points are related to the states with *in-plane* spin polarization perpendicular to the wavevector $(S_u, \text{ positive and negative spins, respectively}).$ Importantly, there is a spin-splitting in the BE region between 4-6 eV, that is, as the momentum approaches to K, in very good agreement with our SR-ARPES data (Figures 5 and 6 and data analysis in Supplementary Information). This splitting is highlighted by red and blue solid lines in Figure 7 and is compatible with the energy spin-splitting determined from the experimental results, that is, we recall, ~ 100 meV. By having a closer look at the Gr π states with higher *in-plane* spin component at Figure 7 (b), they appear to be hybridized with the states localized at Ir (and Co) at the interface (larger DOS inside the rectangle in Figure 7 (a)). Thus, our theoretical calculations largely agree with the experimental SR-ARPES results and demonstrate the strong electronic interaction between electronic states of Gr and those of the Ir-Co interface, as well as an appreciable *in-plane* energy spin-splitting. In other words, the obtained results confirm the Rashba-like spin-texture induced by Ir in the graphene layer.

FIG. 7. (a) DFT band structure for a Gr 1 × 1/Co 2 ML/Ir(111) system along $\overline{\Gamma KM\Gamma}$. The size of the circles is proportional to the product of the Gr and interface Ir DOS, while the color indicates different in-plane spin-polarization in the graphene layer. (b) *In-plane* S_y spin contribution at graphene to the band structure along $\overline{\Gamma K}$ corresponding to the rectangle in panel (a).

CONCLUSIONS

In this work, we have characterized the electronic, spin polarized structure of high-quality $Gr/Co/Ir(111)/Al_2O_3(0001)$ heterostructures with PMA as a function of the Co thickness, by combining ARPES, SR-ARPES experiments and DFT calculations.

Our ARPES results on Gr/Ir(111)/Al₂O₃(0001) reveal a quasi-free standing graphene band structure very similar to the one observed in Gr on Ir(111) single crystals. Upon Co intercalation, the electronic structure is consistent with an epitaxial and two-dimensional intercalation process. In particular, the electronic structure close to E_F for an intercalated layer of 2 ML of Co is very enlightening. On the one hand, the main electronic Ir features at the Co/Ir(111) interface are preserved at E_F , while they become strongly modified just slightly below E_F due to the interaction and hybridization with Co 3d electronic states. On the other hand, at the Gr/Co interface, the Co electronic states strongly interact with C states, resulting in a strong charge transfer to Gr bands and the formation of the mini Dirac cones at the K point because of the Co 3d - C $2p_z$ hybridization. These results are confirmed by DFT calculations. The source of SOC at the Gr/FM interface can be thus attributed to the interaction of the HM layer (Ir) via the FM (Co) intercalation, through an appropriate FM thickness.

In addition, SR-ARPES data and spin-polarized DFT calculations prove the existence of an energy splitting of the *in-plane* spin polarized Gr π bands, consistent with an extrinsic SOC effect, rather a Rashba-like spin-texture induced by Ir in the graphene layer, and that may be related to the DMI experimentally found at Gr/Co [40, 41]. To this concerns, it is interesting to note that the interfacial potential gradient at Gr/Co (i.e., the work function difference between the two layers), first reported in [41], has been very recently correlated to the interfacial effective DMI, as a general behavior for different metal/Co interfaces [60]. Finally, thicker intercalated Co layers have the effect of decoupling Gr from Ir and no spin-splitting is observed. The experiments highlight the role of the HM support by providing the SOC to the entire structure.

METHODS

Growth and preparation methods: The samples were grown at the Molecular Beam Epitaxy (MBE) chamber at IMDEA Nanociencia (Madrid, Spain) and then they were transferred to Cassiopée beamline endstation at SOLEIL Synchrotron (Gif-sur-Yvette, France) via an Ultra-High-Vacuum (UHV) transfer system. The growing method has been described previously by Ajejas *et al.* [49]. In brief, the Gr-based epitaxial heterostructures were grown in UHV conditions on commercially available $Al_2O_3(0001)$ -oriented oxide single crystals. The oxide crystals were ex-situ annealed in air at 1370 K for 2



hours in order to obtain flat surfaces with large terraces prior to their insertion in the UHV chamber. Epitaxial (111)-oriented 30 nm thick Ir buffers were deposited by DC sputtering in 8×10^3 mbar Ar partial pressure at 670 K with a deposition rate of 0.3 Å/s. The quality of the fabricated Ir templates resembles the one of a single crystal, as demonstrated by LEED and XPS surface analyses. The Gr monolayer (ML) was generated on $Ir/Al_2O_3(0001)$ templates by exposing the samples kept at 1025 K in UHV (1 \times 10⁹ mbar) to ethylene gas at a partial pressure of 2×10^8 mbar for 30 min. The Gr/Ir/oxide sample was cooled down to RT and Co was deposited on the top by e-beam evaporation at RT with a deposition rate of 0.04 Å/s. The sample was gradually heated up to 550 K while acquiring XPS spectra to verif in real time the intercalation of Co underneath the Gr sheet. The XPS spectrum of Co was not modified by the presence of CoC [49] which rules out diffusion of C. Once the intercalation had been completed, the resulting sample was $Gr/Co/Ir/Al_2O_3(0001)$.

ARPES experiments: Samples were soft-annealed after the transference to desorb residual gases on the surface prior to ARPES measurements. ARPES were performed at Cassiopée beamline HR-ARPES end-stations. ARPES measurements were performed at a photon energy $h\nu = 64$ eV and the beamline parameters were set to have an energy resolution of 10 meV. ARPES measurements were performed with linear horizontal polarized light and recorded with a Scienta R4000 hemispherical analyzer with an acceptance angle of \pm 15 deg and an angular resolution better than 0.1 deg. ARPES measurements were taken at room temperature (RT), at a base pressure of 10^{-10} mbars.

SR-ARPES experiments: SR-ARPES were taken at the Spin-ARPES end-station of Cassiopée beamline at SOLEIL Synchrotron, at RT and at the same photon energy $h\nu = 64$ eV thanks to an SES2002 Scienta analyzer coupled to a Mini-Mott detector, where the *in-plane* spin polarization component (parallel to the horizontal axis of the surface plane) and *out-of-plane* spin component (perpendicular to the surface plane) were simultaneously recorded. The energy resolution was estimated on 230 meV, while the angular acceptance angle for the channeltrons was over 3.6 deg.

DFT calculations: We used DFT in the generalized gradient approximation [61] employing the full-potential linearized augmented planewave method as implemented in the FLEUR code [62] was applied to investigate the electronic structure of (1×1) Gr/Co(2 ML)/Ir(111). A seven layer Ir(111) film was used as substrate, different stackings were considered and the structures were allowed to relax. The lowest energy was obtained when one carbon atom was situated above the top Co and the other in fcc position. For the band-structures, spin-orbit coupling with an out-of-plane spin-quantization axis was considered, non-collinear calculations allow to detect the

in-plane spin components in the different layers.

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