Emergent magnetism at transition-metal–nanocarbon interfaces

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Charge transfer at metallo–molecular interfaces may be used to design multifunctional hybrids with an emergent magnetization that may offer an eco-friendly and tunable alternative to conventional magnets and devices. Here, we investigate the origin of the magnetism arising at these interfaces by using different techniques to probe 3d and 5d metal films such as Sc, Mn, Cu, and Pt in contact with fullerenes and rf-sputtered carbon layers. These systems exhibit small anisotropy and coercivity together with a high Curie point. Low-energy muon spin spectroscopy in Cu and Sc–C₆₀ multilayers show a quick spin depolarization and oscillations attributed to nonuniform local magnetic fields close to the metallo–carbon interface. The hybridization state of the carbon layers plays a crucial role, and we observe an increased magnetization as sp³ orbitals are annealed into sp²– π graphitic states in sputtered carbon/copper multilayers. X-ray magnetic circular dichroism (XMCD) measurements at the carbon K edge of C₆₀ layers in contact with Sc films show spin polarization in the lowest unoccupied molecular orbital (LUMO) and higher π^* -molecular levels, whereas the dichroism in the σ^* -resonances is small or nonexistent. These results support the idea of an interaction mediated via charge transfer from the metal and dz– π hybridization. Thin-film carbon-based magnets may allow for the manipulation of spin ordering at metallic surfaces using electrooptical signals, with potential applications in computing, sensors, and other multifunctional magnetic devices.

emergent magnetism | molecular spintronics | interfacial magnetism | charge transfer | nanocarbon

Interfaces are critical in quantum physics, and therefore we must explore the potential for designer hybrid materials that profit from promising combinatory effects. In particular, the fine-tuning of spin polarization at metallo–organic interfaces opens a realm of possibilities, from the direct applications in molecular spintronics and thin-film magnetism to biomedical imaging or quantum computing. This interaction at the surface can control the spin polarization in magnetic field sensors, generate magnetization spin-filtering effects in nonmagnetic electrodes, or even give rise to a spontaneous spin ordering in nonmagnetic elements such as diamagnetic copper and paramagnetic manganese (1–11).

The impact of carbon-based molecules on adjacent ferromagnets is not limited to spin filtering and electronic transport, but extends to induced changes in the metal anisotropy, magnetization, coercivity, and bias (12–14). Charge transfer and $d(\text{metal})$ – $\pi(\text{carbon})$ orbital coupling at the interface may change the density of states, spin population, and exchange of metallo–carbon interfaces (4, 15, 16). The interaction between the molecule and the metal depends strongly on the morphology and specific molecular geometry (17, 18). It may lead to a change in the density of states at the Fermi energy DOS(E_F) and/or the

exchange–correlation integral (I_s) as described by the Stoner criterion for ferromagnetism (19, 20). The coupling can even extend through a nonmagnetic metallic layer that carries the exchange information from a ferromagnetic substrate to an organic layer (21).

The emergent magnetism measured in Cu/ and Mn/C₆₀ multilayers extends as well to other transition metals close to complying with the Stoner criterion such as Sc and Pt (Table 1). We grow our films via dc- or rf sputtering and in situ thermal evaporation (*SI Appendix*, section 1). Although we could expect that metals closer to the Stoner criterion would present higher magnetization, it is apparent that the magnetic properties of the sample are strongly determined by the charge transfer and coupling between the carbon molecules and the metal. This is the case of Sc compared with Cu; although Sc is very close to fulfilling the condition for ferromagnetism, it is also a material very prone to oxidation. Furthermore, the Fermi energy and the structure of Sc are not particularly well matched to those of C₆₀. Copper, on the other hand, has a good dz– π coupling, close lattice matching, and the potential to transfer up to three electrons per fullerene cage, and this may result in the higher observed magnetization. The fullerenes may also be replaced by other nanocarbon allotropes with mixed sp² and sp³ hybridization, such as rf-sputtered amorphous carbon (aC) layers. These films have the advantages of being smoother, cheaper than fullerenes, and easily compatible with conventional metal sputtering, making them more suitable for potential industrial applications

Significance

Interfaces are critical in quantum physics, and therefore we must explore the potential for designer hybrid materials that profit from promising combinatory effects. In particular, the fine-tuning of spin polarization at metallo–organic interfaces opens a realm of possibilities, from the direct applications in molecular spintronics and thin-film magnetism to biomedical imaging or quantum computing. This interaction at the surface can control the spin polarization in magnetic field sensors, generate magnetization spin-filtering effects in nonmagnetic electrodes, or even give rise to a spontaneous spin ordering in nonmagnetic elements such as diamagnetic copper and paramagnetic manganese.

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Table 1. Magnetic properties of different nanocarbon hybrids with 2–3-nm-thick metal layers and the structure Si(substrate)/Ta(3)/nanocarbon/metal/nanocarbon

System	Magnetization, emu/cc metal	Coercivity, Oe
Cu/C ₆₀	67 ± 10	90 ± 5
Sc/C ₆₀	17 ± 7	75 ± 10
Mn/C ₆₀	18 ± 2	75 ± 5
Pt/C ₆₀	14 ± 4	65 ± 10
Cu/aC	35 ± 5	95 ± 10
Sc/aC	14 ± 7	75 ± 5

All materials are deposited in the same chamber without breaking vacuum and with a base pressure $\leq 2 \times 10^{-8}$ mbar. Metals are grown via dc magnetron sputtering, C₆₀ is thermally evaporated, and amorphous carbon (aC) is rf-sputtered from a graphite target.

in the future. However, the resulting magnetization as measured using a superconducting quantum interference device (SQUID)–vibrating sample magnetometer (SI Appendix, section 1) is on average 20–40% lower than when using C₆₀, although aC-based samples preserve the same trend observed in C₆₀ multilayers, i.e., higher magnetization and coercivity when using copper films.

The magnetization of Cu and Mn–C₆₀ multilayers has been measured to be dependent on the metallic film thickness, with a peak at 2–5 nm. However, the magnetic moment in Sc/C₆₀ samples is roughly constant. For aC/Cu samples, a factor of 2 higher

moment is measured between ~ 2 –5 nm, but this difference is not as large as for Cu/C₆₀, where the moment is a factor of 6 higher for Cu layers 2–2.5-nm thick than for layers of 3–4 nm (4). This would be expected if the magnetic contribution is mostly due to the interfacial region, resulting in a surface magnetization of ~ 0.05 emu/m² in C₆₀/Sc and 0.1 emu/m² in aC/Cu (see fits in Fig. 1A). The presence of a magnetic moment for films below ~ 1 nm in Sc/C₆₀ seems indicative of a good wetting of the C₆₀ by the Sc film, leading to continuous films or large superparamagnetic islands with high susceptibility at room temperature. Copper films grown on C₆₀, on the other hand, tend to form islands that fill the rifts formed on the uneven molecular film (rms roughness of ~ 1 –2 nm compared with ≤ 0.5 nm for sputtered metals) before forming a continuous layer. The effect of the metal film thickness in the total roughness of the hybrid multilayer is also related to the formation of clusters. As seen in Fig. 1B, samples with thin metal layers are smoother than equivalent samples with thick metal layers, which may have an impact on the magnetization.

Pt and Sc–C₆₀ samples result in a relatively weak magnetization and low coercivity H_c . According to the Stoner–Wohlfarth model (22, 23), the coercive field in the easy axis, H_c , should be equal to $2K/M_s$, where K is the magnetocrystalline uniaxial anisotropy and M_s is the saturation magnetization. For superparamagnetic systems with random crystal orientations, this equation is modified so that H_c drops with temperature until it reaches a zero ideal value for single magnetic domains at the blocking temperature T_B (24):

Fig. 1. (A) Magnetometry in Si/Ta(3)/C₆₀(15)/Sc(t)/C₆₀(15)/Al and Si/Ta(3)/aC(5)/Cu(t)/aC(5)/Al multilayers; lines are fit to a constant interfacial magnetic moment. (B) Sample roughness measured via atomic force microscopy (AFM) vs. number of layers (N) when each film thickness is kept constant (blue dots; line fit to N^2) or when the total multilayer thickness is kept constant (green squares; line fit to asymptotic decay). Both curves should intercept at $n = 4$, but different AFM tips shift them with respect to each other by 1–2 Å. (C) ZFC-FC characteristic measured at 100 Oe for a 0.2- and a 2-nm Sc sample normalized to the low-temperature FC moment. The measurement shows thermal hysteresis typical of systems such as superparamagnets and spin glasses. (D) The coercivity of thin Sc films with C₆₀ increases upon cooling following the dependence for superparamagnetic systems but with a high blocking temperature of 885 K. (E) C₆₀/Sc(2)/C₆₀ samples have a high saturation field that increases at low temperatures and with fields perpendicular to the sample. (F) The simulated contribution of the carbon atoms (M_C) to the total magnetic moment (M_{tot}) for different carbon allotropes and densities shows that the magnetism in the carbon layers is more significant for aC/Cu than for C₆₀/Cu. The as-sputtered aC has a density of ~ 1.7 g/cc, whereas for annealed graphitic films it is ~ 2.3 g/cc. (G) The simulations predict as well a larger Stoner product $I \times \text{DOS}(E_F)$ for C₆₀/Cu than for most aC/Cu systems, in line with the experiments that show larger moments in C₆₀–metal samples (Table 1).

$$H_C \approx \frac{K}{M_s} \left[1 - \frac{T}{T_B} \right]. \quad [1]$$

Zero-field cooled–field-cooled (ZFC-FC) measurements of thin Sc multilayers (<1 nm) show a thermal hysteresis not characteristic of ferromagnetic materials, but rather of superparamagnets or spin glasses below their blocking temperature (Fig. 1C) (25). The magnetic anisotropy derived from the fit of the coercive field to Eq. 1 is very small, $\lesssim 500 \text{ J/m}^3$ —similar to that of permalloy (Fig. 1D). On the other hand, the blocking temperature is very high, some 885 K, corresponding to a grain size of 20 nm ($K \cdot V \approx T_B$). Thicker Sc(2 nm)/C₆₀ films display shape anisotropy with saturation fields of 5 kOe (10 kOe) at 300 K and 12 kOe (17 kOe) at 2 K for in-plane fields (out-of-plane), respectively. The sample saturation magnetization at low temperatures is not reached until fields of >10 kOe are applied for both in-plane and out-of-plane measurements. The atomically rough interface may give rise to weakly coupled magnetic moments with random orientation that do not align easily in smaller fields. That can lead to the observed behavior at low temperatures and the ZFC-FC measurements for ultrathin films (<2 nm) with enough random dipolar fields.

Density-functional theory simulations show that the magnetization is more concentrated at the interface for aC/Cu than for C₆₀/Cu samples, resulting in a slower decay of the interface magnetic moment with the metal layer thickness (*SI Appendix, section 2*). This effect is linked to a higher magnetic contribution of the carbon

layers in the case of aC compared with C_{60} (Fig. 1F). Up to 95% of the moment in Cu/ C_{60} is computed to be in the metal, with the remaining 5% in the C_{60} , but for Cu/aC the moment in the carbon can be as high as 40% of the total. The calculated Stoner product of the exchange integral (Is) with the density of states at the Fermi level [DOS(E_F)] is also smaller for aC/Cu than for C_{60} /Cu, which agrees with the lower measured magnetization (Fig. 1G).

To investigate the magnetic order at the interface and the propagation length of the effect, we use low-energy muon spin rotation (μ SR) to provide a magnetic profile of the sample (26–29). Here, a beam of almost fully polarized positive muons (μ^+) is moderated to kiloelectronvolt (keV) energies so that their tunable stopping range is on the order of tens to hundreds of nanometers. The time evolution of the muon spin polarization is a highly sensitive probe of the local magnetism. This is measured through the detection of the anisotropically emitted decay positrons, preferentially emitted along the muon’s spin direction at the moment of the decay (see [SI Appendix, section 2](#) for further detail). We use this technique to probe two Sc- C_{60} and Cu- C_{60} samples whose structure includes a thin Sc film of 2 nm (2.5 nm for Cu- C_{60}) and a thick Sc film of 5 nm (15 nm for Cu- C_{60}). Nanoscaling of nonmagnetic metals has been shown to give rise to spin ordering (30–33). The thin films should give rise to a strong magnetic signal, whereas for the thicker films, the interfacial magnetization should be quenched or diluted by the bulk properties of the metal. Full sample structures and muon stopping profiles are shown in Fig. 2*A* and *B*.

The muon spin spectroscopy measurements in Sc-C₆₀ and Cu-C₆₀ multilayers show similar properties. At 250 K, both samples

Fig. 2. Muon stopping profile for (A) multilayer with the structure Si/Ta (3)/ C_{60} (20)/ Sc (2)/ C_{60} (50)/ Sc (5)/ C_{60} (20)/ Au (10) ($\text{Sc}-\text{C}_{60}$) and (B) multilayer with the structure Si/Ta (3)/ C_{60} (25)/ Cu (2.5)/ C_{60} (50)/ Cu (15)/ C_{60} (25)/ Au (25) ($\text{Cu}-\text{C}_{60}$); film thickness in brackets in nanometers. All measurements at 250 K unless otherwise indicated. The amplitude of the slowly relaxing component in the muon precession is reduced as we approach the thin metal films in (C) $\text{Sc}-\text{C}_{60}$ and (D) $\text{Cu}-\text{C}_{60}$, indicating the presence of a depolarization term. Lines are fits to the percentage of muons implanted at each layer contributing to the decay. Schematics show this contribution for each multilayer in a red color scale. In remanence, the decay is faster but the contributions remain proportionally the same. The magnetic contribution of the bottom metal- C_{60} interfaces leads to a drop of the muon decay asymmetry at 10–12 keV ($\text{Sc}-\text{C}_{60}$) or 18 keV ($\text{Cu}-\text{C}_{60}$). (E, *Top*) Muon oscillation amplitude at zero field in $\text{Sc}-\text{C}_{60}$ (1.1 MHz). (*Bottom*) In remanence with two contributions (1.1 and 0.8 MHz). Lines are a fit to the layers contribution, with the bottom metal and C_{60} layers having the largest input. (F) Oscillation amplitude (*Top*) and frequency (*Bottom*) for the muon precession in $\text{Sc}-\text{C}_{60}$ with an applied transverse field. Muonium oscillation intensity (1.2, 7.8, and 9 MHz) at 20 K compared with the percentage of muons implanted in C_{60} layers for (G) $\text{Sc}-\text{C}_{60}$ and (H) $\text{Cu}-\text{C}_{60}$. Local magnetic fields at the bottom layers reduce the muonium signal.

have an exponential decay of the polarization at zero field and at remanence that may be due to the presence of local magnetic fields with dynamic or topological variations (34, 35). The amplitude of this signal can be modeled as a function of the contribution of each individual layer in the samples. We find that the C_{60} interfaces with thin metal layers have a much stronger contribution to the depolarization; e.g., for the Sc- C_{60} sample only 2% of the muons stopped in the top C_{60} layer (in contact with Au and thick Sc) contribute to the fast exponential decay vs. 60% of those stopping in the bottom C_{60} layer close to the thin Sc film; see boxes in Fig. 2 C and D. Similar fast exponential decay terms can be observed in antiferromagnetic or ferrimagnetic systems with spin canting, which would explain the low remanent magnetization observed in these samples.

The presence of local hyperfine fields may also be evidenced by a zero-field precession signal at 250 K of ~ 1.1 MHz in Sc/ C_{60} and ~ 0.4 MHz in Cu/ C_{60} . Due to the lack of screening electrons in the molecular semiconductor, muons couple with electrons in C_{60} to form the bound-state muonium (μ^+e^-) (36, 37). Two muonium states are known in C_{60} : (i) the exohedral muonium radical with anisotropic hyperfine coupling which is observable below 100 K where the C_{60} rotational modes are frozen, and (ii) the endohedral muonium state with a large (vacuum muonium-like), isotropic hyperfine coupling. At 250 K the zero-field precession of the exohedral muonium state is not observable due to the fast C_{60} rotation. The isotropic endohedral muonium state should not cause a zero-field precession in the MHz range. However, charge transfer from the transition metal to the C_{60} may cause a deformation of the cage as it is observed for the C_{60} spin triplet state (38). This deformation may result in a slightly anisotropic hyperfine coupling of the endohedral muonium state, which then gives rise to the observed precession frequency in the megahertz range, similar as has been observed in C_{70} (39).

The difference in the zero-field precession frequencies could then be related to different charge and spin states as a function of the transition metal used as a substrate. The frequencies of oscillation shift in remanence after an applied magnetic field of 300 Oe, and the amplitude of the signal is once again stronger when probing the bottom Sc or Cu/ C_{60} interface (Fig. 2E). Fig. 2F (Top) shows the change in the precession amplitude attributed to field inhomogeneities. This can be related as well to the random orientation of local dipoles in small fields due to film roughness. The oscillation frequency of the muons in an applied transverse external field of 140 Oe is also increased for implantation energies corresponding to thin Cu layer due to the additional local field (Fig. 2F, Bottom).

Below 100 K, it is possible to observe the zero-field muonium precession frequencies of the exohedral radical state in the range of 1–9 MHz. The fact that the muonium oscillation frequencies are clearly observable implies that the magnetization must be localized close to the Cu layers; otherwise the internal fields would shift/destroy these modes in C_{60} . Their amplitude can be used to probe the magnetic properties of the sample at 20 K (Fig. 2 G and H). Given the number of particles implanted in C_{60} , the muonium signal should increase to reach a maxima at 8 keV (Sc- C_{60}) or at 18 keV (Cu- C_{60}). However, the signal remains roughly constant, indicative of magnetic fields perturbing the muonium signal at the higher implantation energies toward the bottom layers.

The orbital hybridization and molecular coupling with the metal are essential to the charge transfer and emergent magnetism. C_{60} films degrade in ambient conditions due to light-induced oxidation, which leads to a decay of the magnetization in a few days if the structures are not protected (Fig. 3A). The concentration of oxygen in C_{60} exposed to atmospheric conditions drops when heated in vacuum at 400 K, reaching near-pristine levels by 450 K (40). The magnetism at Sc/ C_{60} interfaces follows this deoxygenation trend, with increased magnetization after

Fig. 3. Degradation, annealing, and carbon hybridization effects. (A) Time decay of the magnetization in Cu/ C_{60} multilayers. In samples with several layers, the bottom interfaces are protected from chemical degradation by the top layers, and the magnetization decay is slow. (B) By annealing, the magnetization of a Si/ C_{60} (15)/Sc(3nm)/ C_{60} (15) sample can be enhanced or the degradation compensated. (C) Raman spectra of rf-sputtered amorphous carbon on Cu after annealing. At 475–775 K, the G peak moves to higher frequencies and decreases in intensity relative to the D peak. At 875 K and above, the G peak increases again and the D peak has a replica at 1,275 cm^{-1} . (D) Evolution of the magnetization and carbon structures during the annealing process. Lines are a guide to the eye. Schematics show the sample structure changes derived from the Raman spectra.

heating to 400–500 K. At higher temperatures, the C_{60} is desorbed from the metallic substrate, leading to a reduced magnetization (Fig. 3B).

For rf-sputtered carbon, annealing alters the orbital states, bonding, and structure of the layers. We use Raman spectroscopy to track these changes via two vibrational modes: the D band due to the breathing of benzene rings, and the G band due to the stretching of the C–C bond (E_{2g} mode at the Γ -point) (41, 42). As the annealing temperature is increased, the G band shifts to higher frequencies, from 1,520 to 1,615 cm^{-1} . The intensity of the D band (I_D) with respect to the G band (I_G) gets progressively higher; I_D/I_G increases from ~ 0.6 in as-deposited films to ~ 2 after annealing at 875 K for 1 h (Fig. 3C). The Raman changes are evidence for the conversion of amorphous carbon into nanocrystalline graphite and the change of orbital hybridization from sp^3 to sp^2 (41–43). In as-deposited films, the estimated percentage of sp^3 orbitals from the I_D/I_G ratio and G-peak position is ~ 15 –20%, but at 875 K the percentage has dropped to zero. Above 875 K, graphitic nanocrystals expand until they form full graphite sheets. This leads to the G peak shifting back to lower frequencies and the I_D/I_G ratio decreasing again. In addition to these well-characterized changes, above 875 K there is the formation of a new peak at 1,275 cm^{-1} that we have labeled as D^* . This peak is at a frequency below those conventionally assigned to the D band in graphite for a 532-nm source even in strain/stressed samples (1,340–1,380 cm^{-1}) (44). This shift could be due to charge transfer and reduction of the carbon atoms.

The structural changes result in a reduced magnetization but increased coercivity (Fig. 3D), with a net enhancement in the $B \times H$ energy product (*SI Appendix, section 3*). These results are reproducible for samples annealed in situ (i.e., heated inside the SQUID at 50 mtorr He atmosphere using an oven probe) and ex situ (annealed in an external oven under vacuum and then remeasured). The solubility of carbon in solid Cu above 1,100 K for 72 h—i.e., at higher temperatures and for longer times than those used in our experiment—is below 5 atomic ppm (45). Although the solubility of rf-sputtered aC in thin films of Cu could be different from that in bulk, we do not observe any sign of interdiffusion.

An independent measurement of the magnetization contribution of the carbon material is provided by soft X-ray absorption spectroscopy (XAS), exploiting its inherent chemical specificity when the X-ray photon energy is tuned at the carbon K-edge electronic transition. It can therefore be used to assess the presence of magnetic ordering in a given element separately from the contribution of other layers or impurities using X-ray magnetic circular dichroism (XMCD) (46). The K-edge XMCD can only probe orbital polarization due to the zero angular momentum of the 1s core shell (47, 48). In Fig. 4 we present results in the near-edge X-ray fine absorption spectroscopy (NEXAFS) and XMCD at the carbon K edge of a Sc- C_{60} sample under a 1-T applied magnetic field along the X-ray beam direction, incident at a 45° angle on the film. The edge structure of C_{60} on the metal surface is complex, and at room temperature it includes features at 284 (lowest unoccupied molecular orbital, LUMO), 286.2, 286.7, and 287.5 eV (excited LUMO+1,+2,+3) in the π^* -antibonding region (49, 50). The positions of these peaks are lower in energy than for pure C_{60} , which has previously been observed for fullerene films in contact with other conducting magnetic substrates (51, 52). The σ^* -region is above the ionization potential and the modes are less defined, producing wider peaks. The highest unoccupied molecular orbital–LUMO transition ($h1u-t1u$) in C_{60} is optically forbidden, although it is weakly present via vibrational excitations.

In XMCD spectra at room temperature, the first unoccupied states at the LUMO ($t1u-t1g$ levels) appear as magnetically ordered with the positive X-ray polarization having lower electron yield current—we refer to this as negative dichroism, observed here in the absence of magnetic material. Conversely, the following

Fig. 4. X-ray spectroscopy of C_{60} grown on a Sc 4-nm film collected at room temperature (Top) and at 2 K (Bottom). The measurements are done under an applied 1-T perpendicular magnetic field in the total electron yield mode (TEY) and a 45° beam incidence. The NEXAFS show the typical structure of C_{60} -suggested modes in the bottom panel. Both measurements show magnetic dichroism, indicating the presence of a considerably large orbital polarization in the C_{60} density of states—even in the absence of a magnetic substrate. Note that XMCD measurements at low temperatures are multiplied by a smaller factor and that there is an additional, negatively polarized peak provisionally assigned to an SAMO or CEX.

peaks around 287.5 eV (LUMO+2,+3) have positive dichroism. In contrast to this spin polarization for π^* -states, the σ^* -antibond orbitals are weakly or not polarized (Fig. 4). Similar results with a negatively polarized LUMO and positive higher π^* -states can be obtained in samples with other Sc film thicknesses and under other experimental configurations (*SI Appendix, section 4*). The measurements show a time dependence due to charging effects and/or radiolysis, but the sign of the dichroism is changed when the X-ray polarization is reversed or a negative magnetic field is applied.

There is a small peak some 5.5 eV above the LUMO, at 289.5 eV. This energy is too high for a charged LUMO π^* -state. However, the resonance is too narrow and well-defined for a σ^* -state. Also, the peak becomes much stronger at low temperatures, indicating a quantum state difficult to observe at 300 K. We hypothesize that these characteristics may be interpreted as due to a delocalized superatom molecular orbital (SAMO), previously measured using low-temperature scanning tunneling microscopy in C_{60} deposited on a metal surface (53). The highest relative photoionization cross-section would correspond to the s-wave orbital (54), but here the energy gap to the LUMO is closer to values observed in microscopy for p- or d-wave orbitals. Another possibility is that this peak constitutes a carbon core exciton (CEX) close to the ionization potential (55, 56). XMCD measurements show this state to have the same polarization direction as the LUMO. In addition to this prospective magnetically polarized SAMO, two nonpolarized states become visible at low temperatures: the metal-coupled charged carbon state at 282–283 eV, and the peak due to aggregated or graphite-like C_{60} at 285.3 eV.

In conclusion, we have shown evidence for the universality of the emergent magnetism in metal–nanocarbon interfaces and its magnitude in different materials. Our results give evidence that it is indeed possible to have spin-polarized states at a metallic interface with molecular carbon even in the absence of magnetic materials. This is of critical importance in the design and measurement of organic spintronic devices and magnetic field sensors, where these interfaces can be used as spin filters. Furthermore, given changes in polarization at the different energy levels, a gate voltage may give us tuning access to spin up/down configurations or new quantum configurations, such as spin-polarized superatom orbitals or polarons. The possibility of tailoring the magnetic properties of transition metal–nanocarbon hybrids by using molecular interfaces opens as well tantalizing possibilities—for example, in

noncorrosive magnets, bio-compatible hybrid nanoparticles, metal recovery and in magnetic memories where the information is controlled via charge transfer with electrooptic irradiation (5, 6).

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