Organic spintronics is an emerging research topic in the last two decades due to exciting physical phenomena as well as its potential in various spintronic applications. Organic semiconductors (OSCs) have drawn immense research interest for spintronic applications due to low spin orbit coupling, less hyperfine interaction, long spin lifetime, low cost and mechanical flexibility [1–4]. Spin valve like structure using OSC as a spacer layer has already been shown to exhibit high magnetoresistance [5–8]. It has been reported that the performance of the spin valve device depends on the interface of the OSC and ferromagnet (FM) [3, 4, 9–15]. There is a high chance of formation of a spinterface due to spin polarized charge transfer and orbital hybridization at the OSC-FM interface [16, 17]. Because of this, the density of states of the OSC gets modified and the OSC can exhibit ferromagnetism [10, 18–25].

The effect of spinterface has been shown in many in-plane magnetized systems. Recently, we have shown that due to the spinterface the magnetization reversal and domains are modified in an epitaxial MgO (100)/Fe/C_{60} system [20]. The magnetic moment per C_{60} cage was found to be \( \sim 2.95 \mu_B \) [20]. Similarly, for C_{60} deposited on a polycrystalline Fe films also exhibited spinterface and domain size was reduced [22]. Further in another study, we have shown that the spinterface in a Co/C_{60} system has similar effect, however the anisotropy increased due to the spinterface [23]. Although there have been a few works on in-plane magnetized films, however, the study of such spinterface on domains in a perpendicularly magnetized films are scarce.

Perpendicular magnetic anisotropic (PMA) systems are the most suitable candidates for data storage devices due to their high intrinsic anisotropy [26]. Due to high thermal stability and low energy consumption, the PMA based devices have advantages over the in-plane ones. In systems with PMA, magnetic moments are aligned perpendicular to the film plane which enhances the spin-flipping efficiency thereby reducing the required current density which may be useful for spin-orbit torque (SOT) based devices [26, 27]. Bairagi et al. demonstrated that with increasing the thickness of the OSC (C_{60}) layer on a Co ultrathin film one can tune the anisotropy of the system from in-plane to out-of-plane [28, 29]. It has been also shown that a C_{60} layer can enhance the PMA of the Ni thin film in a Ni/C_{60} bilayer system [30]. Further, it has been observed that adsorbing the organic molecules on CoFe3N surface enhance the PMA of the system [31].

Therefore, the study of domain and magnetic relaxation is quite appealing in FM-OSC systems with PMA. From application point of view, domain engineering via various approaches are appealing. For example, recently we have shown that by making magnetic antidot lattices (MALs) in a Pt/Co/Pt film, the domain size was significantly reduced [32]. However, domain engineering via spinterface and in particular in Pt/Co/Pt system has not been studied so far. In this paper, we have considered a Pt/Co/Pt system which exhibits high PMA. We have studied a Pt/Co/C_{60}/Pt system in which the effect of spinterface at the Co-C_{60} interface on magnetic domain size and relaxation have been investigated. The formation of spinterface and its effect on the local magnetic environment has been studied by means of density functional theory (DFT) calculations. We have observed that due to the spinterface the size of bubble domains gets significantly reduced. Further from the magnetization relaxation measurements it is found that the relaxation time decreases for the Pt/Co/C_{60}/Pt system.

We have prepared two Pt/Co/Pt thin films with and
FIG. 1. (a) hysteresis loop measured by MOKE based microscopy in polar mode for sample 1. (b - e) are the corresponding domain images at magnetic fields of 50, -24.73, -26.89 and -27.85 mT, respectively.

FIG. 2. (a) hysteresis loop measured by MOKE based microscopy in polar mode for sample 2. (b - e) are the corresponding domain images at magnetic fields of -50, -11.72, -12.10 and -12.30 mT, respectively.

without C\textsubscript{60}. The sample structures are the following:
Sample 1: Si/ Ta (5.5 nm)/Pt (4.1 nm)/Co (0.8 nm)/Pt (4.6 nm) and
Sample 2: Si/ Ta (5.5 nm)/Pt (4.1 nm)/Co (0.8 nm)/C\textsubscript{60} (1.7 nm)/Pt (4.6 nm)
The thin films have been prepared using DC sputtering (Co, Ta), RF sputtering (Pt) and thermal evaporation (C\textsubscript{60}) techniques in a multi-deposition high vacuum chamber manufactured by Mantis Deposition Ltd., UK. The base pressure in the chamber was $5 \times 10^{-8}$ mbar. All the layers were deposited in-situ to avoid surface contamination and oxidation. Ta was used as a seed layer to promote the (111) direction growth of Pt. Further to maintain the growth uniformity, we have rotated the substrate at 20 rpm during deposition of all the layers. To prevent from oxidation of the films a Pt capping layer of 4.6 nm thickness was deposited. We have measured the magnetic domains and relaxations at room temperature by magneto-optic Kerr effect (MOKE) based microscopy manufactured by Evico magnetics GmbH, Germany. The measurements were performed in polar mode where the magnetization is parallel to the plane of incidence and
FIG. 3. (a) Relaxation data for sample 1 measured at $H_M=0.97 \ H_C$, where the solid circles represent the raw data and red line is the best fit using compressed exponential function given as equation (1). (b) - (e) represent the domain images captured at 0, 4, 8, 17 sec respectively. The scale bar shown in (b) is same for all the images and the applied field is out of the plane.

FIG. 4. (a) Relaxation data for sample 2 measured at $H_M=0.97 \ H_C$, where the solid circles are representing the raw data and red line is the best fit using compressed exponential function given in equation (1). (b) - (e) represent the domain images captured at 0, 2, 4, 17 sec respectively. The scale bar shown in (b) is same for all the images and the applied field is out of the plane.

perpendicular to the sample surface.

The density functional theory (DFT) calculations were performed using Vienna Ab-initio Simulation Package (VASP) [33] in order to understand the atomistic details and the electronic structure of the spinterface. In the calculations the valence electronic states are expanded with a plane wave basis set, while the core electrons are treated with pseudopotential. The valence-core interaction was represented by full-potential Projected Augmented Wave (PAW) method [34]. The Generalized Gradient Approximations (GGA) is used to treat the exchange-correlation potentials with the Perdew, Bruke, and Ernzerof (PBE) functional [35]. A plane-wave cut-off of 500 eV was used to guarantee a good convergence of the total energy. The convergence tolerance for the self-consistent electronic minimization was set to $10^{-8}$ eV/cycle. For optimiza-
A Gaussian smearing parameter of 0.2 eV is used to smear the bands. A Monkhorst-Pack K-points grid is taken as (3×3×1) for the surface slabs. A vacuum of more than 10 Å was added to the z-direction of the surface super cells to ensure no interaction between periodic images at that particular direction. The Co 3d orbitals are treated with a $U_{eff}=3.0$ eV for the calculation of magnetic anisotropy energy (MAE) and exchange coupling. The surface slabs were constructed using optimized cell parameters $a=2.50$ Å and $c/a=1.6$ for hexagonal closed packed (HCP) Co and $a=2.48$ for face centered cubic (FCC) Co.

The thickness of the Co layer is very crucial for PMA system. In order to know the exact thickness and roughness of all the layers, we have performed X-ray reflectivity (XRR) measurement and fitted the data using GenX software. The XRR data and their best fits are shown in supplementary information figure S1. It has been observed...
that the thickness of the Co layer is same for both the Pt/Co/Pt and Pt/Co/C_{60}/Pt samples (refer to the Table S1 in supplementary information). Figure 1 (a) shows the hysteresis loop measured by MOKE based microscopy in polar mode for sample 1. The coercive field ($H_C$) is 25.56 mT. Figure 1 (b-e) show the domain images of sample 1 at positive saturation field (+$H_S$), nucleation field ($H_N$), near to coercive field ($H_C$) and negative saturation field (-$H_S$), respectively. Bubble domains are observed in the samples as the anisotropy ratio, $Q = K_u/K_d \gg 1$, where $K_u$ and $K_d$ are perpendicular anisotropy and stray field energy densities, respectively [36]. With increasing the magnetic field bubble domains expand and at saturation field they merge with each other. Figure 2 (a) shows hysteresis loop and figure (b-d) show the domain images of sample 2 at +$H_S$, $H_N$, $H_C$, and -$H_S$, respectively. It is clearly observed that the size of the domains for sample 2 become smaller in comparison to sample 1. Also, the coercivity for sample 2 (11.55 mT) is less than that of sample 1. The possible reason behind it may be the formation of spininterface at Co-C_{60} interface which increases the anisotropy as well as the decrease of domain size of the system. However, the overall nature of the bubble domains remains same like its parent Pt/Co/Pt thin film.

Depending on the domain dynamics, switching speed of spintronic devices can be tuned. To calculate the relaxation time of a system we have performed magnetization relaxation measurement using Kerr microscopy at room temperature. Here first we have saturated the sample and then reversed the magnetic field to a sub-coercive field (0.93 $H_C$, 0.95 $H_C$ etc.) and kept the field constant. The magnetization will relax with time under a constant Zeeman energy and complete the reversal process via domain nucleation and/or domain wall (DW) motion under the influence of thermal activation energy. Domain images were captured in a regular interval of time during this process.

We have calculated average normalized intensity using ImageJ software and plotted it with respect to time. The relaxation curve can be fitted using various models [37–42]. Here we have fitted our experimental data using compressed exponential function [32]:

$$I(t) = I_1 + I_2 (1 - \exp\left(-\frac{t}{\tau}\right)^{\beta}) \tag{1}$$

where $I(t)$ represents the Kerr intensity at time $t$, $I_1 + I_2$ is normalized Kerr intensity measured at saturation, $\tau$ is relaxation time constant and $\beta$ is an exponent having value between 1 (domain nucleation dominated magnetization reversal) and 3 (dominated by DW motion).

Figure 3 (a) shows the relaxation behaviour of Pt/Co/Pt thin film (sample 1) at $H_M$=0.97 $H_C$ and (b – e) show the domain images captured at 0, 4, 8, 16 seconds, respectively, which are marked in (a). Figure 3(b) has been captured at $t = 0$, i.e. just after applying the constant magnetic field ($H_M$). After that domains are evolved with time under the influence of thermal activation energy. From the fitting of the relaxation curve using equation (1), we have found $\beta = 1.640 \pm 0.057$, which ensures that magnetization reversal is dominated by domain nucleation followed by DW motion [32]. Similarly, figure 4 (a) shows the relaxation behaviour of Pt/Co/C_{60}/Pt thin film (sample 2) at $H_M$=0.97 $H_C$ and (b – e) show the domain images captured at 0, 2, 4, 16 seconds, respectively, which are marked in (a). By fitting the data to equation (1), the $\beta$ value is found to be $1.120 \pm 0.061$ which also indicates domain nucleation dominated magnetization reversal[32].

| Sample name | 0.97Hc | 0.95Hc | 0.93Hc |
|-------------|-------|-------|-------|
|              | $\tau$ | $\beta$ | $\tau$ | $\beta$ | $\tau$ | $\beta$ |
| Sample 1    | 5.28 ± 0.08 | 1.64 ± 0.06 | 5.62 ± 0.04 | 1.53 ± 0.02 | 10.45 ± 0.14 | 1.67 ± 0.05 |
| Sample 2    | 2.80 ± 0.10 | 1.12 ± 0.06 | 3.34 ± 0.16 | 1.22 ± 0.09 | 5.48 ± 0.10 | 1.42 ± 0.05 |

FIG. 7. The charge density reorganization at the C_{60}/Co interface. Cyan indicates the depletion of charge density while yellow isosurface indicates the accumulations.

To study the speed of the relaxation with different Zeeman energy, we have performed the relaxation mea-

TABLE I. Parameters obtained from the best fits of the data shown in Fig. 5 using compressed exponential function equation (1)
TABLE II. Parameters used to define the interface formation between different Co-slabs and C$_{60}$

| System               | Penta- C$_{60}$ | E$_{ad}$(kcal/mol)$^{a}$ | e$^{-}$-transferred$^{b}$ | Number of Co-C bond$^{c}$ |
|----------------------|-----------------|--------------------------|----------------------------|---------------------------|
| HCP-Co(0001)         |                 | -94.89                   | 1.50                       | 9                         |
| Penta-Hexa- C$_{60}$ |                 | -100.30                  | 1.41                       | 8                         |
| FCC-Co(111)          |                 | -83.15                   | 1.45                       | 7                         |
| Penta-Hexa C$_{60}$  |                 | -95.10                   | 1.39                       | 8                         |

$^{a}$ Adsortion Energy, $E_{ad}=E_{Co+C_{60}}-(E_{Co}+E_{C_{60}})$
$^{b}$ e$^{-}$ - transferred from Co substrate to C$_{60}$ = Bader population on C$_{60}$ in Co+C$_{60}$ - Bader population on C$_{60}$.
$^{c}$ Co-C bond distance Cut-off = 2.2 Å

TABLE III. Computed relative energies of the antiferromagnetic (AFM-i) spin configuration where one magnetic moment is antiferromagnetically coupled with the rest of the substrate atoms. $\Delta E$ has been calculated as $\Delta E = E_{AFM-i} - E_{FM}$. For surface all the magnetic moments are identical and hence only one relative energy is there. The MAE has been calculated here, as MAE = E$_{axis}$-E$_{Z-axis}$. Positive MAE value indicates out of plane magnetic anisotropy.

| System               | MAE(meV) | $\Delta E$(meV)$^{*}$  |
|----------------------|----------|------------------------|
|                      | X-axis   | Y-axis | AFM-1(1.9) | AFM-2(1.8) | AFM-3(1.6) |
| HCP-Co(0001)          | 8.25     | 8.26   | 661.44     |            |            |
| +Penta-C$_{60}$       | 7.55     | 7.37   | 935.06     | 698.16     | 626.39     |
| +Penta-Hexa-C$_{60}$  | 8.94     | 8.78   | 916.15     | 660.50     | 717.04     |
| FCC-Co(111)           | 0.59     | 0.58   | 642.33     |            |            |
| +Penta-C$_{60}$       | 0.91     | 0.63   | 928.64     | 673.75     | 694.67     |
| +Penta-Hexa-C$_{60}$  | 2.00     | 1.98   | 914.67     | 673.90     | 686.08     |

* Values inside the parenthesis represent the approximate magnetic moments in $\mu_{B}$ (DFT+U) of the corresponding ferromagnetic configuration which has been inversed in a particular AFM configuration.

measurements at different sub-coercive fields (0.93, 0.95, 0.97 $H_C$) for both the samples 1 and 2. Figure 5 (a) and (b) show the relaxation behaviour for samples 1 and 2, respectively, measured at $H_M = 0.97$ $H_C$ (blue squares), $H_M = 0.95$ $H_C$ (red triangles), $H_M = 0.93$ $H_C$ (green circles). The $\beta$ and $\tau$ values obtained from the best fits to equation (1) for both the samples are given in Table 1. For both the samples relaxation time ($\tau$) decreases with increasing the applied magnetic field, which is expected. However, for Pt/Co/C$_{60}$/Pt thin film (sample 2) the relaxation time is reduced by $\sim 50\%$ as compared to Pt/Co/Pt thin film (sample 1). Therefore, it is inferred that by introducing a C$_{60}$ layer the switching speed of a system can be tuned which is a promising way to build device applications.

The nature of the spinterface at the Co-C$_{60}$ interface has been explored applying DFT calculations. Two different types of Co-substrates are considered here; viz, HCP-Co (0001) and FCC-Co (111), as the exact morphology of the experimental samples are not known. Two different sites of C$_{60}$ were deposited on the Co-substrates terming them as ‘pentagonal’ and ‘pentagonal-hexagonal’ configurations. The formation of such spin-interface amicably modifies the isotropic magnetic exchange interactions of the surface atoms as well as amends the magnetic anisotropy of the substrates. The quantitative estimations of such interfacial magnetic properties are obtained from our first principle-based computations.

Formation of the spin-polarized interface is shown in Figures 6, where the C$_{60}$ is inversely spin polarized as compared to the Co-substrate. Figure 7 depicts how the electron density is getting redistributed due to formation of spinterface. From dml-resolved density of state (DOS) (Figure 8) of the surface Co atoms, it is evident that the $d_{(2z)}$ and $d_{x}$ orbitals form a new hybridized orbital at the bonding site. On the other hand, the C-atom directly attached to the Co atom changes it’s hybridization from $sp^2$ to $sp^3$ (Figure 9.). These changes of the hybridization of the atoms at the spinterface leads to the formation of Co-C bond (Figure 10) and as a consequence spin split states are formed at the non-magnetic C atoms. A comparison of different spinterface related properties is given in Table 2. Penta-Hexa adsorption of C$_{60}$ leads to more stable configuration.

There are mainly three types of magnetic moment present at the interface Co layer (Supplementary information figures S2 and S3). We have considered three different antiferromagnetic (AFM-i) spin configuration where the spin moment of one of the atoms with a particular magnetic moment is reversed. The energy difference between the corresponding antiferromagnetic configuration and the ground state ferromagnetic configuration indicates how the local magnetic exchange interactions are being modified due to the formation of the spinterface as compared to the clean surface. For most of the configurations the magnetic exchange interactions are enhanced. This reveals that the local magnetic environment at the adsorption site become discrete from rest of the ferromag-
The magnetic anisotropy for the interfaced systems is calculated accounting the spin-orbit couplings in the non-collinear magnetic calculations. The magnetic easy-axis is aligned out of plane i.e, perpendicular to surface layers for all the Co-surfaces studied here. The values of the magnetic anisotropy energy (MAE) w.r.t. to the easy axis anisotropy for pristine Co surface as well as for C$_{60}$ adsorbed surface are compared in Table 2. It is evident that there are enhancements in MAE (except for penta-C$_{60}$ on HCP-Co which is less favorable configuration) due to the adsorption of the C$_{60}$ on the Co-substrate. This indicates the hardening the pinning of out-of-plane magnetization due to strong adsorption of C$_{60}$ on Co-substrate. In our previous study on Co-C$_{60}$ in-plane system, we have also seen that introducing a C$_{60}$ layer anisotropy of the system has been increased [23].

The magnetization relaxation and domain wall dynamics in perpendicular magnetic anisotropic thin films of Pt/Co/Pt with and without C$_{60}$ have been discussed in this paper. Bubble domains are observed for both the thin films. However, introducing the C$_{60}$ layer in Pt/Co/Pt thin film reduces the size of the domains. The C$_{60}$-substrate interactions not only modifies on-surface isotropic as well as anisotropic magnetic textures compared to the bare substrates but it causes the reduction...
in domain size. The engineering in domain size is advantageous for memory storage application point of view. Similarly, the speed of magnetic relaxation (τ value) is faster for Pt/Co/C₆₀/Pt sample. This fast switching has a remarkable impact in device application. Further in this Pt/Co/C₆₀/Pt tri-layer structure it is expected that a finite amount of interfacial Dzyaloshinskii–Moriya interaction (iDMI) may be observed. In future the quantification of iDMI may be performed to explore the possibility of tuning the iDMI to host any chiral magnetic structures.

The authors would like to thank Dr. Tanmay Chabri for technical help during measurement. We also thank Dr. Sougata Mallick and Dr. Braj Bhusan Singh for valuable discussions. We acknowledge Department of Atomic Energy (DAE), and Department of Science and Technology - Science and Engineering Research Board, Govt. of India (DST/EMR/2016/007725, CRG/2019/003237) for the financial support.

Author contributions

SB has conceived the idea and coordinated the project. SB has designed the experiments and MEA has designed the DFT calculations. SM has prepared the samples. PS and SM have performed the Kerr microscopy experiments. AM has done the DFT calculations. All authors have discussed the results and written the manuscript.

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Supplementary Information
Effect of Fullerene on domain size and relaxation in a perpendicularly magnetized Pt/Co/C$_{60}$/Pt system

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Structural information
In order to investigate the structural information, we have performed X-ray reflectivity (XRR) measurements by a x-ray diffractometer (model SmartLab) manufactured by Rigaku. We have fitted the data by using GenX software. Figure S1 (a), (b) show the XRR data and best fits for samples 1 and 2, respectively. Structural parameters such as thickness and roughness of the layers are shown in Table S1.

Distribution of spin moment
Figures S2 and S3 depict how the spin moments are distributed over different atomic sites at different Co-C$_{60}$ interface. The top layer of the Co slabs is affected mostly by the spinterface formation. There are mainly three types of spin moments at the considered interfaces. These moments have been used to calculate the exchange interactions.

FIG. S1. XRR data and the best fits for samples 1 and 2 are shown in (a) and (b), respectively. The open circles are experimental data and the solid lines represent the best fit using GenX software. The parameters extracted from the best fits are shown in Table S1.

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arXiv:2012.12777v2 [cond-mat.mtrl-sci] 25 Dec 2020
TABLE I. Parameters obtained from XRR fits

| Layers | Sample 1 |           | Sample 2 |           |
|--------|----------|-----------|----------|-----------|
|        | Thickness (nm) | Roughness (nm) | Thickness (nm) | Roughness (nm) |
| Ta     | 5.8      | 0.75      | 5.5      | 0.56      |
| Pt     | 4.5      | 0.74      | 4.1      | 0.57      |
| Co     | 0.8      | 0.50      | 0.81      | 0.49      |
| C60    | -        | -         | 1.7      | 0.77      |
| Pt     | 4.8      | 0.698     | 4.6      | 0.69      |

FIG. S2. Distribution of spin moments on different atomic sites for HCP-Co (0001) substrate.

FIG. S3. Distribution of spin moments on different atomic sites for FCC-Co (111) substrate.