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Boosting magnetoresistance in manganese phosphide helimagnets through iron oxide interface strain engineering

Nivarthana W. Y. A. Y. Mudiyansele,^a Derick DeTellem,^a Yasinthara M. Wadumesthri,^a Amit Chanda,^b Anh Tuan Duong,^c Emmanuel Olawale,^a Humberto Rodriguez Gutierrez,^d Xiaomei Jiang,^a Sarath Witanachchi^a and Manh-Huong Phan^{a,d}

Helimagnetic materials exhibit complex non-collinear spin structures, making them promising candidates for next-generation magnetoresistive devices. Their unique magnetic textures can yield enhanced magnetoresistance effects compared to conventional materials, potentially improving the performance of magnetic sensors and memory technologies. In this study, an interface engineering strategy is presented that leverages strain induced by the structural phase transition of Fe₃O₄ to enhance the magnetoresistance (MR) effect in nanostructured manganese phosphide (MnP) films near the ferromagnetic-to-helimagnetic (FM–HM) transition. MnP films (~100 nm thick, grain size ~86 nm) were grown on Si substrates using molecular beam epitaxy, followed by deposition of Fe₃O₄ layers of varying thicknesses (7 nm and 58 nm). The results show that a thin Fe₃O₄ layer (7 nm) enhances interfacial magnetic coupling and overall magnetization in the bilayer, while the thicker Fe₃O₄ layer (58 nm) dominates the magnetic response due to its soft magnetic nature. Remarkably, the MR near the FM–HM transition increases by 20% and 37% with the 7 nm and 58 nm Fe₃O₄ layers, respectively, attributed to strain-enhanced spin-dependent scattering at the interface. These findings provide new insights into strain-modulated magnetic coupling at iron oxide/helimagnet interfaces and underscore their potential for advanced spintronic applications.

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Introduction

Helimagnetism is a type of magnetic order characterized by a non-collinear arrangement of spins, resulting in a helical or spiral structure.^{1–3} In helimagnets, the magnetic moments of the atoms rotate continuously, forming a helix along a specific axis. This configuration can support the formation of topological spin textures, such as skyrmions.^{4,5} This contrasts with ferromagnetism, where spins are aligned parallel, and antiferromagnetism, where they align antiparallel. Helimagnetism presents a rich area of study in condensed matter physics, with

implications for both fundamental research and practical applications.^{6–10}

Helimagnets can be utilized in spintronic devices, which exploit the spin of electrons for information processing and storage, potentially leading to faster and more efficient technologies.^{11–14} Their unique magnetic configurations may enhance data storage solutions, allowing for higher density and faster access times in hard drives and memory devices.² Additionally, helimagnets can be employed in highly sensitive magnetic sensors, benefiting applications in industrial, automotive, and consumer electronics.

Magnetoresistance (MR) is a key principle in spintronic devices, leveraging both the charge and spin of electrons to create more efficient electronic components, including memory and logic circuits.¹⁵ Materials exhibiting large MR are crucial for developing energy-efficient devices, which is particularly important for reducing power consumption in electronic applications. Investigating magnetoresistance in helimagnets opens new avenues for research and applications, making it a critical area in materials science and condensed matter physics.^{16–18} The unique magnetic properties of heli-

^aDepartment of Physics, University of South Florida, Tampa, Florida 33620, USA.
E-mail: nivarthanawa@usf.edu, phanm@usf.edu

^bDepartment of Energy Conversion and Storage, Technical University of Denmark, 2800 Kgs. Lyngby, Denmark

^cFaculty of Materials Science and Engineering, Phenikaa University, Hanoi 12116, Vietnam

^dCenter for Materials Innovation and Technology, VinUniversity, Hanoi 100000, Vietnam



magnets have been predicted to result in enhanced MR effects in the helimagnetic regime compared to ferromagnetic materials,¹⁹ potentially enhancing the efficiency and sensitivity of sensors and memory devices.

Helimagnetic phenomena have been observed in a variety of magnetic systems, such as MnSi,²⁰ MnP,^{21,22} MnGe,²³ MnAuS₂,¹⁴ DyTe₃,²⁴ CrI₂,²⁵ Cr_{1/3}NbS₂,^{26–28} Fe_xCo_{1–x}Si²⁹ and FeGe.³⁰ However, a comprehensive understanding of their properties has remained elusive. Among these, manganese phosphide (MnP) has garnered significant attention due to its multifunctional magnetic, superconductive, thermoelectric, magnetocaloric, and magnetoresistive properties.^{13,31–34} Bulk MnP exhibits a complex magnetic phase transition, transitioning from paramagnetic (PM) to ferromagnetic (FM) at $T_C \approx 291$ K, followed by an FM to helimagnetic (HM) transition at $T_N \approx 47$ K.³⁴ Nanostructuring has been reported to considerably alter the HM behavior in MnP, with notable increases in the FM–HM transition temperature (up to 110 K) in nanocrystalline films.^{22,35} The unique magnetic behavior of MnP, driven by the Dzyaloshinskii–Moriya interaction (DMI), makes it a promising candidate for emergent magnetism and magneto-transport in heterostructure systems.

In a recent study, we discovered a large magnetoresistance effect in MnP nanostructured films near the FM–HM transition by leveraging confinement, strain effects, and spin helicity.³⁶ This highlights a novel strain-mediated spin helicity phenomenon in nanostructured helimagnets, presenting a promising pathway for developing high-performance magnetoresistive sensors and spintronic devices. Further research into such nanostructured helimagnets and their heterostructures, formed by interfacing MnP with other functional magnetic materials, may lead to new materials with tailored properties for specific applications in electronics and materials science.

In this context, iron oxide (Fe₃O₄) emerges as an excellent candidate for forming heterostructures with MnP. Fe₃O₄ possesses an inverse spinel crystal structure, where O^{2–} ions are arranged in a cubic close-packed manner.^{37–39} The unit cell features two distinct cation sites, referred to as A and B sites. A sites are occupied by Fe(III) ions, forming tetrahedral coordination, while B sites contain equal numbers of Fe(II) and Fe(III) ions in octahedral coordination. This structure allows magnetite to be represented as [Fe³⁺]_A[Fe²⁺Fe³⁺]_BO₄. The electron hopping between Fe³⁺ and Fe²⁺ at the B sites enhances conductivity compared to other iron oxides like Fe₂O₃ and FeO.^{37,38} Furthermore, the spins of Fe³⁺ at A sites and those of Fe²⁺ and Fe³⁺ at B sites are antiferromagnetically coupled *via* superexchange, giving Fe₃O₄ its ferrimagnetic properties.³⁷ Fe₃O₄ also exhibits a high Curie temperature (~860 K) and high spin polarization (~100%), making it desirable for spintronics applications.⁴⁰ At room temperature, Fe₃O₄ behaves as a semiconductor, transitioning to an insulating state around 120 K *via* the Verwey transition, which is accompanied by a structural transformation from cubic inverse spinel to a monoclinic phase.^{37,38,41} A substantial body of research has focused on the magnetic, multiferroic, magnetoresistive, and spin-thermo-

transport properties of Fe₃O₄ films and heterostructures, which aim to explore them for applications in spintronics and spin-caloritronics.^{40,42–47}

The formation of Fe₃O₄/MnP interfaces in bilayer structures is expected to lead to novel magnetic and magneto-transport phenomena due to proximity and exchange coupling effects, as well as structural modifications at the interface.^{40,42,46} Structural changes around the Verwey transition of Fe₃O₄ may significantly impact the Fe₃O₄/MnP interface coupling and the higher MR behavior of MnP nanostructured films near the FM–HM transition temperature. Using techniques such as X-ray diffraction (XRD), atomic force microscopy (AFM), magnetic force microscopy (MFM), and temperature- and magnetic field-dependent magnetometry and resistivity, our study sheds light on the emergent magnetic and magnetoresistance phenomena in Fe₃O₄/MnP heterostructures. We demonstrate that the presence of a Fe₃O₄ layer can increase the MR effect by 13% in Fe₃O₄/MnP bilayers, with the MR ratio tunable by varying Fe₃O₄ thickness. By introducing the “universal curve” analytical method, we show that the intrinsic transport properties of MnP in the helimagnetic state are consistent across samples, regardless of Fe₃O₄ thickness. Our findings pave the way for developing novel heterostructures based on nanostructured helimagnets with enhanced MR properties, meeting the increasing demands of modern spintronic and sensor devices.

Results and discussion

Structural characterization

Since the structural properties of the MnP films used in this study were thoroughly characterized in our previous work,³⁶ we present here those of Fe₃O₄ layers (thickness, ~7 and ~58 nm) deposited on top of the MnP films (average grain size of ~86 nm, film thickness ~100 nm) and Fe₃O₄/MnP bilayers. For thickness determination, part of the MnP film was masked during PLD growth of Fe₃O₄ to leave a bare reference region of MnP. The AFM step-height across this boundary was measured and the resultant profile is shown in Fig. S1. Presence of Fe and O on MnP films were investigated by EDS. Fig. S2 shows the obtained EDS spectra for Fe₃O₄/MnP bilayers. Fig. 1a shows the XRD pattern of the Fe₃O₄ (58 nm)/MnP bilayer, with reference to those of the MnP and Fe₃O₄ films. We observed the characteristic peaks of Fe₃O₄ phase, specifically the (1 1 1), (3 1 1), (2 2 2), and (5 1 1) reflections, which are consistent with the Fe₃O₄ films grown on Si substrates. Additionally, the Fe₃O₄ (58 nm)/MnP film displays (4 0 0) of Fe₃O₄ phase, which is absent in Fe₃O₄ (62 nm)/Si film. Several diffraction planes of MnP phase are present in the Fe₃O₄ (58 nm)/MnP bilayer, confirming that the MnP retains its orthorhombic structure (*Pnma* space group) with lattice parameters of $a = 5.259$ Å, $b = 3.173$ Å, and $c = 5.917$ Å. Both Fe₃O₄ films, deposited on Si and MnP, exhibit a face-centered cubic lattice structure with *Fd* $\bar{3}m$ and *F* $\bar{4}3m$ space groups. The lattice parameters for Fe₃O₄ (62 nm)/Si and Fe₃O₄ (58 nm)/MnP/Si are $a = 8.3963$ Å and $a =$



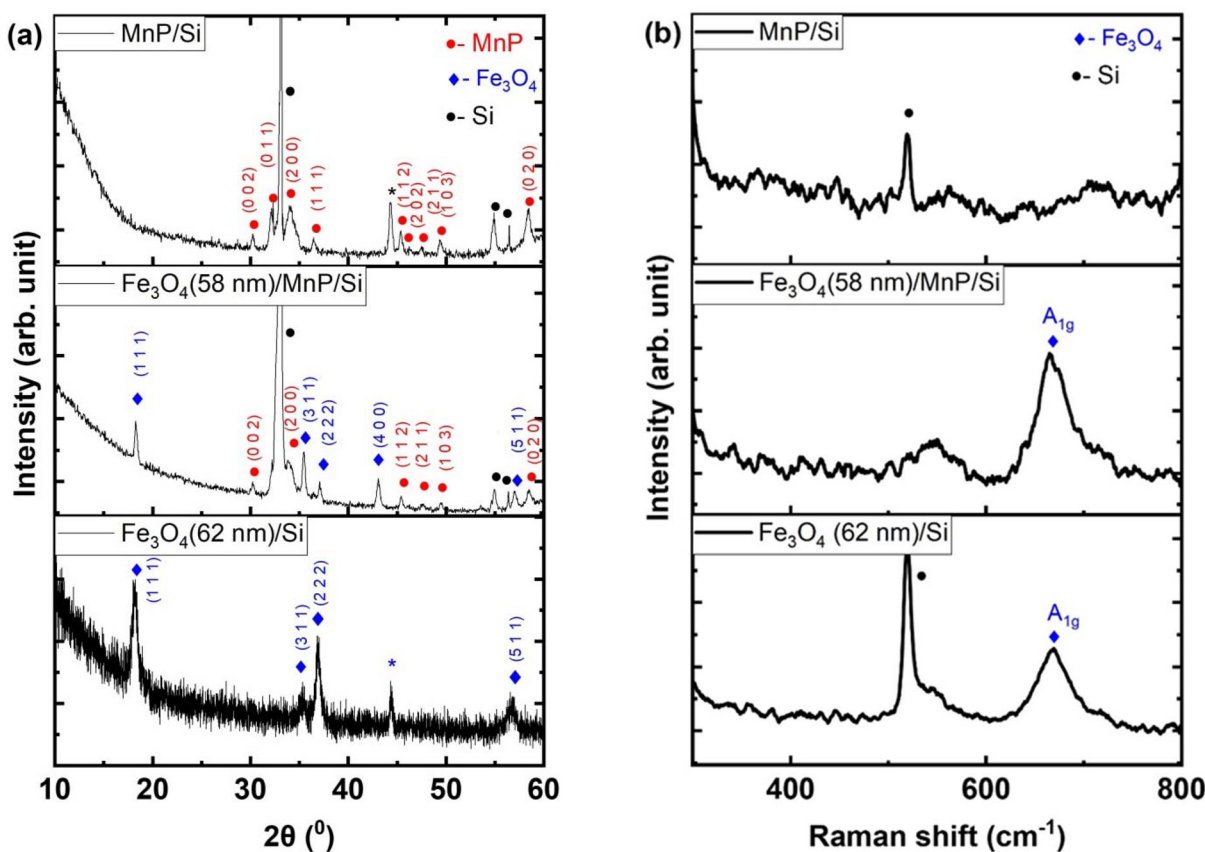


Fig. 1 (a) X-ray diffraction (XRD) patterns of MnP/Si, Fe₃O₄ (58 nm)/MnP, and Fe₃O₄ (62 nm)/Si. The asterisk mark (black) indicates the XRD signal from the sample holder and the asterisk (blue) is due to Si substrate. (b) Raman spectra of MnP/Si, Fe₃O₄ (58 nm)/MnP, and Fe₃O₄ (62 nm)/Si.

8.3941 Å, respectively. This indicates that the substrate plays a critical role in the growth of Fe₃O₄ thin films.^{37,48} Fig. 1b shows the room temperature Raman spectra of MnP/Si, Fe₃O₄ (58 nm)/MnP/Si, and Fe₃O₄ (62 nm)/Si samples. Raman active modes of A_{1g} of Fe₃O₄ in the Fe₃O₄/MnP bilayers and Fe₃O₄/Si are observed at 667 cm⁻¹ (668 cm⁻¹). The peak positions of the Raman spectra are basically consistent with the previously reported data of Fe₃O₄ films.^{49–51} No additional iron oxide phases were detected in the Raman spectra.

Topography images of Fe₃O₄ (7 nm)/MnP and Fe₃O₄ (58 nm)/MnP are displayed in Fig. 2(a and b). The root mean square (RMS) roughness was determined to be 6.1 nm and 3.4 nm, respectively, for the 7 nm and 58 nm Fe₃O₄ films on MnP. The bare MnP film exhibited a roughness of 6.5 nm. Luo *et al.* have shown the correlation between the thickness of graphene and the surface morphology of the substrate when depositing graphene on Cu using chemical vapor deposition method.⁵² Notably, in our study the intended thickness of this deposition was ~80 nm based on the calibrated thickness of Fe₃O₄ on Si substrate (~83 nm), even using the exact same deposition conditions. This discrepancy suggests a difference in the growth mechanism of Fe₃O₄ on MnP compared to that on Si. This substrate roughness of MnP (6.5 nm) could explain the deviation in film thickness from the calibrated thickness,

as the Fe₃O₄ (58 nm)/MnP film exhibits roughly half the roughness of the MnP film. This suggests that during initial nucleation and growth, the Fe₃O₄ fills voids on the MnP surface. Conversely, Fe₃O₄ on Si showed a significantly lower roughness of 0.1 nm. Fe₃O₄ reference films were also deposited on Si substrates. The target thicknesses were chosen to match as closely as possible the Fe₃O₄ layers grown on MnP (7 nm and 58 nm). Due to minor variations in PLD chamber conditions and uncertainties in thickness measurement, the resulting Fe₃O₄/Si reference films were ~10 nm and ~62 nm. These samples are reported throughout the manuscript as Fe₃O₄/Si references. It is generally accepted that the grain size of a thin film grows when the film thickness increases.⁵³ Our Fe₃O₄/Si films also demonstrated this, as indicated by the AFM topography images, Fig. S3(a–c). In contrast, the Fe₃O₄ (7 nm)/MnP/Si shows a random size distribution of grain sizes from large to small (inset of Fig. 2a and Fig. S4a) when compared to Fe₃O₄ (58 nm)/MnP/Si (inset of Fig. 2b and Fig. S4b).

Magnetic force microscopy analysis

MFM measurements were conducted on all samples at room temperature, including Fe₃O₄ (10 nm and 62 nm) on Si, MnP (100 nm) on Si, and their Fe₃O₄/MnP bilayers. Fig. 2(c and d) presents MFM images of Fe₃O₄ (58 nm)/MnP/Si, revealing



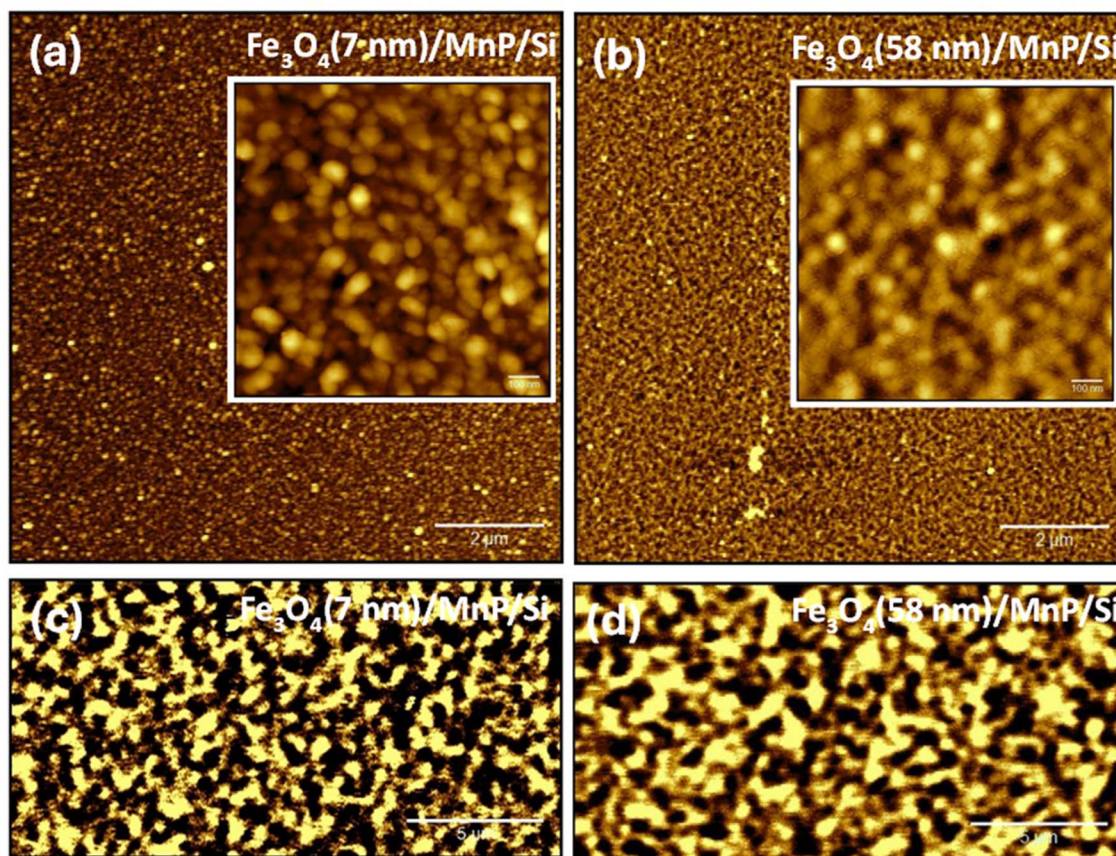


Fig. 2 Atomic force microscopy (AFM) topography images of (a) Fe_3O_4 (7 nm)/MnP/Si and (b) Fe_3O_4 (58 nm)/MnP/Si; room temperature magnetic force microscopy (MFM) images of (c) Fe_3O_4 (7 nm)/MnP/Si and (d) Fe_3O_4 (58 nm)/MnP/Si.

bright and dark contrasts that indicate the presence of randomly distributed magnetic domains. These domains are aligned perpendicularly to the plane of the film. The domain distribution of Fe_3O_4 (58 nm)/MnP/Si is comparable to that observed in the Fe_3O_4 (62 nm)/Si film (Fig. S5a). Similar magnetic domain patterns in Fe_3O_4 films grown on $\text{SrTiO}_3\text{:Nb}$ using PLD were reported by Wei *et al.*⁵⁴ In MFM, the contrast in the domain image arises from the magnetic force gradient $\left(\frac{dF}{dz}\right)$ between the sample and the MFM tip, which is proportional to the stray field of the film.⁵⁵ The root-mean-square (RMS) value of phase shift ($\Delta\phi_{\text{RMS}}$) is defined as $\Delta\phi_{\text{RMS}} = \frac{Q}{K} \left(\frac{dF}{dz}\right)$, where Q is the quality factor and K is the spring constant of the tip.^{56,57} The Q value, K , and lift height were kept constants for all measurements to ensure valid comparisons of the $\Delta\phi_{\text{RMS}}$ values across the bilayers and reference films. At 300 K, the measured $\Delta\phi_{\text{RMS}}$ values were 0.6399 for Fe_3O_4 (7 nm)/MnP, 0.8190 for Fe_3O_4 (58 nm)/MnP, 0.3532 for MnP/Si, 1.1160 for Fe_3O_4 (62 nm)/Si, and 0.1160 for Fe_3O_4 (10 nm)/Si (Fig. S5d). These variations in $\Delta\phi_{\text{RMS}}$ reflect differences in the $\frac{dF}{dz}$ and consequently, the underlying magnetic domain structures and stray field strength. Notably, the

thinner layer (7 nm) of Fe_3O_4 on MnP shows a higher $\Delta\phi_{\text{RMS}}$ value when compared to the Fe_3O_4 (10 nm) film grown on Si (0.1160), indicating that MnP influences the magnetic properties of the Fe_3O_4 layer in Fe_3O_4 /MnP.⁵⁸

Since MFM is a surface-sensitive technique, the magnetic contrast and RMS phase shift values primarily reflect the magnetic properties of the uppermost layers, with the Fe_3O_4 top layer dominating the signal in the Fe_3O_4 /MnP bilayer samples. Comparing the Fe_3O_4 /MnP bilayers with the bare MnP film shows a stronger magnetic signal due to significant stray fields of Fe_3O_4 . When comparing the Fe_3O_4 /MnP bilayers to Fe_3O_4 /Si films, differences in $\Delta\phi_{\text{RMS}}$ values arise from the distinct growth behaviors on each substrate. Growth of Fe_3O_4 on MnP introduces unique magnetic domain structures, possibly due to strain, crystallographic orientation, or interfacial exchange coupling. These effects are less significant in Fe_3O_4 films grown on Si, where the magnetic domains follow a more typical Fe_3O_4 pattern (Fig. S5b) when compared to Fe_3O_4 on MnP (Fig. 2c). The MnP substrate likely alters the Fe_3O_4 growth process, affecting domain size, or magnetic anisotropy, which in turn impacts $\Delta\phi_{\text{RMS}}$.⁵⁸ The influence of sital-substrate roughness on Ta_2O_5 film growth mechanisms was studied by Sergeev *et al.*, showing that the roughness of sital-substrates could lead to different sizes of grains and increased



intergranular stress.⁴⁸ Rougher substrates may also reduce the thickness of the upper layer by enhancing surface diffusion and atom migration, leading to the filling of valleys and smoothing of peaks. This explains the deviation in thickness of the Fe₃O₄ films grown on Si and MnP. The rough surface of MnP significantly influenced the growth of the Fe₃O₄ (7 nm) film and its resulting magnetic properties, in contrast to the Fe₃O₄ (10 nm) film grown on a smooth Si substrate. The difference in grain size observed in Fe₃O₄ (7 nm)/MnP/Si (inset of Fig. 2a) and its higher $\Delta\phi_{\text{RMS}}$, as compared to Fe₃O₄ (10 nm)/Si (Fig. S3c), further support this conclusion. On another note, the deposition of the Fe₃O₄ (7 nm) layer on the rough surface of MnP may influence the magnetic behavior of the MnP film. A dual magnetic proximity effect, arising from the interactions between the MnP and Fe₃O₄ layers, likely contributed to the observed magnetic properties of the Fe₃O₄/MnP bilayers.^{59–61} In contrast, for the Fe₃O₄ (58 nm)/MnP bilayers, the thicker Fe₃O₄ layer predominated the system's magnetism, leading to similar trends in both $\Delta\phi_{\text{RMS}}$ and M_{S} of the Fe₃O₄ (58 nm)/MnP and Fe₃O₄ (62 nm)/Si configurations.

Magnetometry analysis

To understand the impact of Fe₃O₄ deposition on the magnetic properties of MnP thin films, we conducted systematic temperature- and magnetic field-dependent magnetization measurements on all samples, including Fe₃O₄ (7 nm and 58 nm) on Si, MnP(100 nm) on Si, and their Fe₃O₄/MnP bilayers, using the vibrating sample magnetometry (VSM) option of the physical property measurement system (PPMS). Fig. 3(a and b) shows the temperature dependence of in-plane DC magnetization (M - T) of Fe₃O₄ (7 nm)/MnP/Si and Fe₃O₄ (58 nm)/MnP/Si films measured under zero-field-cooled (ZFC), field-cooled-cooling (FCC), and field-cooled-warming (FCW) protocols in presence of a magnetic field 0.1 T. The magnetization behavior of the Fe₃O₄/MnP bilayers exhibits significant differences in the lower temperature region (<100 K) compared to the bare MnP film (inset of Fig. 3a). Notably, the M - T curves under ZFC indicate the larger magnetization values at 300 K for Fe₃O₄ (7 nm)/MnP/Si (~ 63 emu cm⁻³) and Fe₃O₄ (58 nm)/MnP/Si (~ 315 emu cm⁻³) than for MnP (~ 17 emu cm⁻³). The M_{S} of the three films also follows the same trend of the magnetization at 300 K, as indicated in Fig. S5e. This reinforces the interaction between the two materials in the bilayer structure, which arises from the dual magnetic proximity effect.⁶⁰ The temperature-dependent magnetization trend at higher temperatures (>200 K), continues to align with the bare MnP film. In contrast, the Fe₃O₄ layers impact the magnetization profile of the MnP, particularly at low temperatures (<200 K). The first derivative of the M - T dependence of the Fe₃O₄/MnP bilayers shows that the paramagnetic to ferromagnetic (PM-FM) transition of MnP ($T_{\text{C}} \sim 306$ K) remains nearly unchanged, appearing at ~ 304 K for Fe₃O₄ (7 nm)/MnP and ~ 302 K for Fe₃O₄ (58 nm)/MnP (Fig. S6a). However, the ferromagnetic to helimagnetic (FM-HM) transition of MnP, typically observed around $T_{\text{N}} \sim 102$ K, shifts to lower temperatures (~ 90 K) in the

Fe₃O₄/MnP bilayers. It should be noted that the Verwey temperature (T_{v}), which was observed at approximately 118 K in our Fe₃O₄/Si samples (Fig. S6b), where a noticeable slope change in magnetization occurred in the M - T curve. This value is close to the Verwey transition temperature of bulk Fe₃O₄ ($T_{\text{v}} \sim 120$ K).³⁷ It is important to note that the presence of the Fe₃O₄ layer on the MnP film had a negligible impact on the PM-FM transition of MnP (approximately 300 K) but resulted in a shift of the FM-HM transition to a lower temperature (~ 90 K) (Table S1). In the present bilayer system, however, we find no noticeable shift in the PM-FM transition around 300 K, suggesting that the static strain from the Fe₃O₄ overlayer is insufficient to modify MnP's high- T ordering. By contrast, the structural transformation of Fe₃O₄ during its Verwey transition (~ 110 – 120 K) imposes a transient interfacial strain, which is capable of perturbing the MnP layer and shifting its low-temperature FM-HM transition. Additionally, previous reports have highlighted the varying effects of strain (whether tensile or compressive) on the FM-HM transition of MnP. Choi *et al.* observed an increase in T_{N} (approximately 100 K) in MnP films, attributing this change to strain effects resulting from the lattice mismatch between the epitaxial MnP film and the GaAs substrate.⁶² Similarly, de Andrés *et al.* reported a significant increase in T_{N} from 47 K for bulk MnP to 67 K for MnP films grown on GaP substrates, and 82 K for MnP nanocrystals embedded in GaP epilayers, while the T_{C} values remained largely unchanged.⁶³ They suggested that surface strains induced by tensions in the MnP nanocrystals substantially modify the helimagnetic structure, leading to the observed increase in T_{N} . In contrast, Sun *et al.* found a slight decrease in T_{N} (~ 38 K) for a 2D MnP single crystal compared to its 3D bulk counterpart (~ 47 K).³² Gregg *et al.* reported complete suppression of T_{N} in 20 nm orthorhombic MnP nanorods with their growth direction along the b -axis, suggesting that the screw phase was entirely confined along the hard a -axis.⁶⁴ Hirahara *et al.* demonstrated that applying tensile strain along the a -axis of a MnP single crystal favored antiferromagnetic (AFM) interactions over ferromagnetic (FM) ones, resulting in the increased T_{N} and decreased T_{C} . Conversely, compressive strain along the c -axis produced the opposite effects, yielding lower T_{N} and higher T_{C} values.⁶⁵ In the present study, the increase in T_{N} of the MnP nanocrystalline film (~ 102 K), compared to bulk MnP (~ 47 K), is attributed to surface strains induced by tension resulting from the nanostructuring effect. The deposition of the Fe₃O₄ layer on the MnP film appears to weaken the influence of these surface strains on the FM-HM transition, thereby reducing the T_{N} of the MnP film. Additionally, the magnetic proximity effect induced by the Fe₃O₄ layer could significantly contribute to the interfacial magnetic coupling between the two layers, as indicated by the MFM analysis.

To further clarify this, we measured the magnetic field-dependent magnetization (M - H) curves across a broad temperature range of 10 to 350 K for all samples, including MnP/Si, Fe₃O₄ (10 nm)/Si, Fe₃O₄ (62 nm)/Si, Fe₃O₄ (7 nm)/MnP/Si, and Fe₃O₄ (58 nm)/MnP/Si. A comprehensive analysis of these data



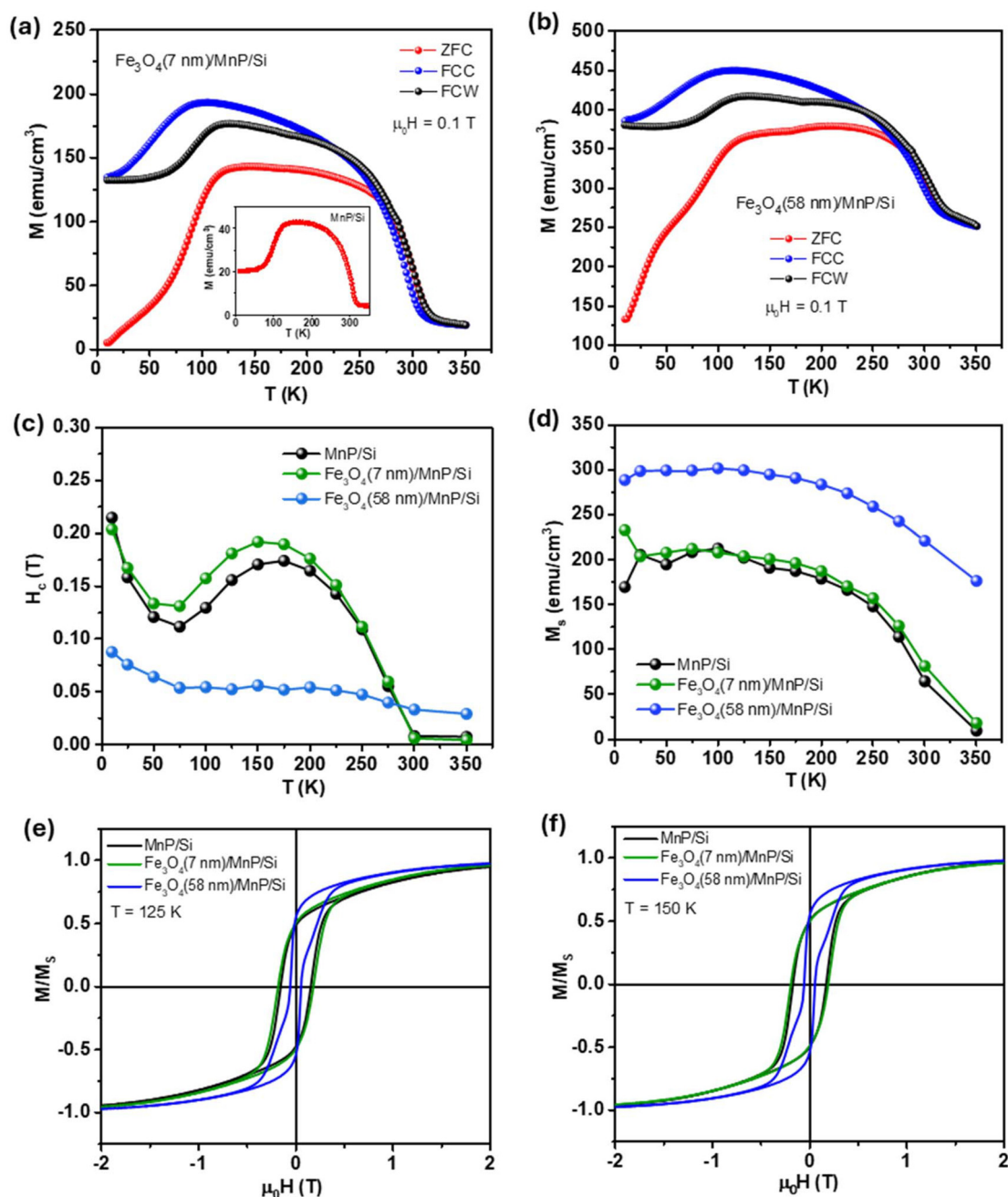


Fig. 3 Temperature dependence of in-plane magnetization (M - T) of (a) Fe_3O_4 (7 nm)/MnP/Si and (b) Fe_3O_4 (58 nm)/MnP/Si. The inset of (a) represents the M - T dependence of MnP/Si under ZFC protocol at 0.1 T; temperature dependence of (c) coercivity (H_c) and (d) saturation magnetization for the three films; magnetic field-dependent magnetization (normalized) for the three films at (e) 125 K and (f) 150 K for the in-plane (IP) configuration.

reveals that the presence of the Fe_3O_4 (7 nm) layer significantly enhanced the coercivity (H_c) of Fe_3O_4 (7 nm)/MnP compared to the bare MnP film, particularly within the temperature range of 50–250 K where the Verwey transition of Fe_3O_4 (~118 K) and the FM-HM transition of MnP (~102 K) occurred; a similar temperature-dependent coercivity trend was observed for both cases though (see Fig. 3c). The temperature-dependent saturation magnetization of Fe_3O_4 (7 nm)/

MnP/Si is comparable with MnP/Si with a small increment throughout the temperature region of 10–350 K (see Fig. 3d). On the other hand, the highest M_s of Fe_3O_4 (58 nm)/MnP/Si supports the prominent effect of Fe_3O_4 when the layer thickness increased. In the temperature regime of interest (50–250 K), the shape of the M - H loops for the Fe_3O_4 (7 nm)/MnP bilayer closely resembles that of the bare MnP film (see Fig. 3(e and f)). These findings suggest that the presence of a



thin layer of Fe_3O_4 enhances magnetic coupling at the $\text{Fe}_3\text{O}_4/\text{MnP}$ interface, contributing to the overall enhanced magnetism of the system (Fig. 3c and d). The situation is notably different for the Fe_3O_4 (58 nm)/MnP bilayer, where the presence of the 58 nm-thick Fe_3O_4 layer, exhibiting soft ferromagnetic characteristics (see Fig. S6(c and d)), dominates the overall magnetism of the system. This results in a significant reduction in H_C and a low-field shrinking loop shape at temperatures below the T_C of MnP (Fig. 3(e and f)).

Transport and magnetoresistance properties

To understand the effect of Fe_3O_4 deposition on the transport and magnetoresistance properties of the MnP films, we conducted temperature and magnetic field-dependent resistivity measurements on the bare MnP film and $\text{Fe}_3\text{O}_4/\text{MnP}$ bilayers over a wide temperature range of 10–300 K. Fig. 4a illustrates the temperature dependence of the longitudinal resistivity (ρ) for MnP/Si, Fe_3O_4 (7 nm)/MnP/Si, and Fe_3O_4 (58 nm)/MnP/Si films measured in zero magnetic field. A clear distinction is observed between the resistivity profiles of the bilayer films and the bare MnP film. The resistivity of MnP exhibits metallic behavior, with a gradual decrease in resistivity as the tempera-

ture decreases to approximately 110 K. The minimum around 97 K, followed by an upturn (maximum at approximately 73 K), is observed and attributed to the confinement and strain effects, as well as magnetic phase coexistence and competition in governing electron transport properties.³⁶ In the Fe_3O_4 -deposited MnP films, this resistivity minimum shifted to lower temperatures, specifically to ~ 85 K for Fe_3O_4 (7 nm)/MnP/Si and ~ 69 K for Fe_3O_4 (58 nm)/MnP/Si. Additionally, the peak of the resistivity upturn, which occurred around 73 K in MnP/Si, shifted to about 61 K for Fe_3O_4 (7 nm)/MnP/Si and 45 K for Fe_3O_4 (58 nm)/MnP/Si. These shifts suggest that the presence of the Fe_3O_4 layer significantly altered the electronic transport properties of MnP, particularly in the low-temperature regime. Guan *et al.* observed the effect of a capping layer (Fe_3O_4) on the metal-insulator transition of $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ films due to the $\text{Fe}_3\text{O}_4/\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ interface effect.⁶⁶ Furthermore, Takahashi *et al.* noted an increase in resistivity with increasing Fe_3O_4 thickness when grown on a metallic spacer layer, Au, attributed to the interface between Fe_3O_4 and the metallic spacer.⁶⁷ In our study, we found that the overall resistivity of the $\text{Fe}_3\text{O}_4/\text{MnP}$ bilayers increased with the thickness of the Fe_3O_4 layer.

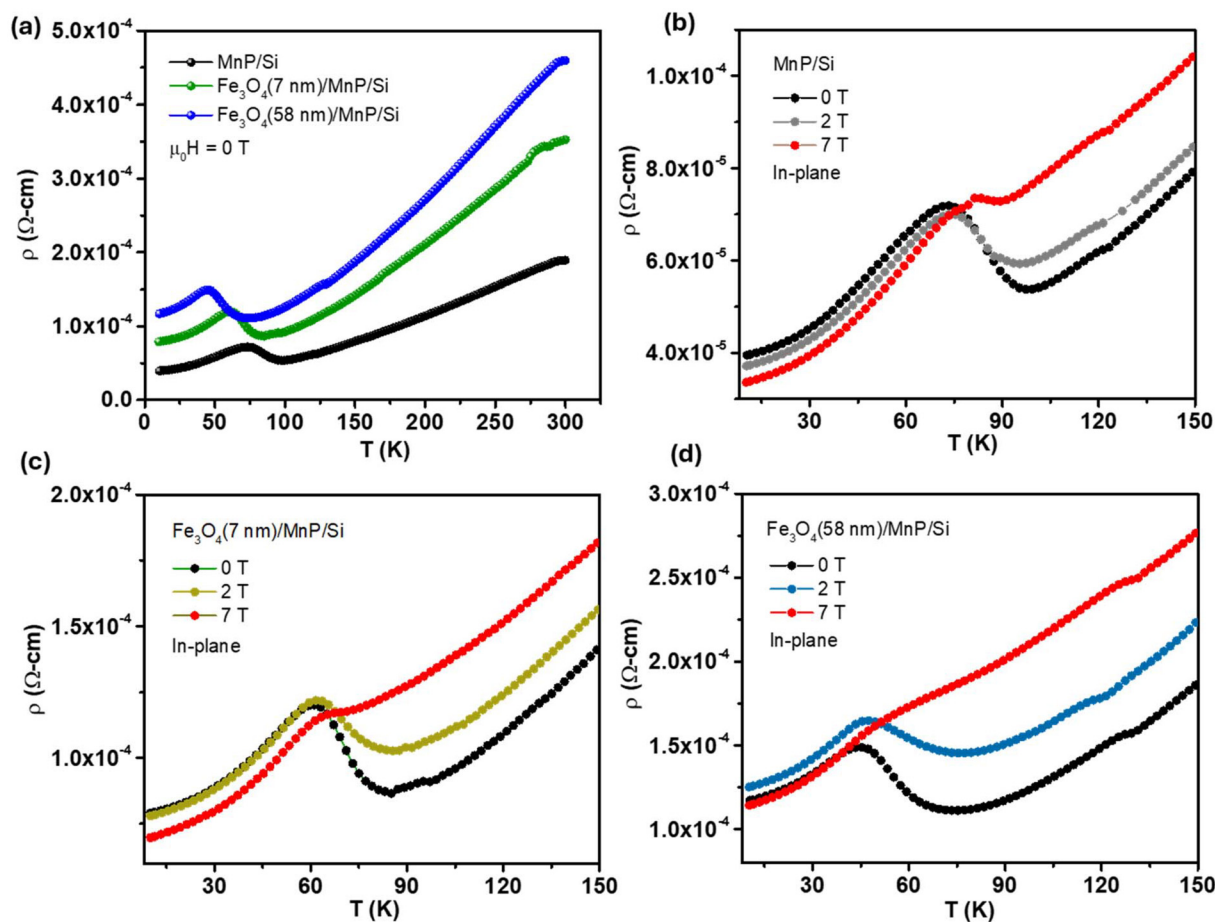


Fig. 4 (a) The temperature-dependent resistivity under zero field for MnP/Si and $\text{Fe}_3\text{O}_4/\text{MnP/Si}$ samples; longitudinal resistivity under in-plane magnetic fields of 0, 2, and 3 T within an enlarged temperature range of 8–150 K for (b) MnP/Si, (c) Fe_3O_4 (7 nm)/MnP/Si, and (d) Fe_3O_4 (58 nm)/MnP/Si.



Fig. 4(b–d) shows $\rho(T)$ under magnetic fields of 0, 2, and 7 T across a temperature range of 10–150 K for MnP, Fe_3O_4 (7 nm)/MnP/Si, and Fe_3O_4 (58 nm)/MnP/Si. It is interesting to note that for the bare MnP film, the application of magnetic fields enhanced the resistivity at temperatures above approximately 75 K, while reducing it at lower temperatures (Fig. 4b). This effect becomes even more pronounced with the application of higher magnetic fields. A similar trend is observed for the Fe_3O_4 (7 nm)/MnP bilayer (Fig. 4c), although the effects are more pronounced due to the presence of the deposited Fe_3O_4 (7 nm) layer. The case is different for the Fe_3O_4 (58 nm)/MnP bilayer (Fig. 4d), where the effects of applied low (2 T) and high (7 T) magnetic fields on resistivity differ significantly. The application of a 2 T field increased the resistivity across the entire temperature range of 10 to 300 K. In contrast, the 7 T field raised the resistivity at temperatures above 50 K but reduced it at lower temperatures. This indicates that the magnetic fields have varying impacts on the magneto-transport properties of Fe_3O_4 /MnP bilayers depending on the thickness of the Fe_3O_4 layer.

To further clarify this intriguing feature, we have performed a thorough analysis of the MR data and display in Fig. 5 the magnetic field-dependent MR profiles of the Fe_3O_4 /MnP bilayers, with reference to the MR profile of the bare MnP film

(Fig. S7). It can be observed that the Fe_3O_4 (7 nm)/MnP bilayer shows a sign change of MR from positive to negative at ~ 65 K (see Fig. 5a and 6a), while the Fe_3O_4 (58 nm)/MnP bilayer exhibits this sign change at ~ 50 K (see Fig. 5b and 6a). Such changes in MR sign resemble those of the bare MnP film, where the MR sign change occurred at approximately 75 K (Fig. S7). This sign change in the MnP film has been attributed to the confinement and strain effects within the FM regime of the MnP nanostructured film, which govern the transport properties and lead to the positive MR behavior in this region.³⁶ The MR magnitude reaches its maximum at approximately 102 K, which corresponds to the FM–HM transition temperature of the MnP film (Fig. 6). The spin-dependent scattering mechanisms at the grain boundaries in the MnP polycrystalline film are thought to be responsible for the positive MR effect. It is noteworthy that the presence of the Fe_3O_4 layer significantly enhances the positive MR effect of the bare MnP film, with the magnitude of MR increasing as the thickness of the Fe_3O_4 layer increases. The maximum MR ratio of the bare MnP film increases by 20% with the 7 nm Fe_3O_4 layer and by 37% with the 58 nm Fe_3O_4 layer in the Fe_3O_4 /MnP bilayers. This enhancement in MR is attributed to proximity-enhanced spin-dependent scattering at the Fe_3O_4 /MnP interface.⁴⁰ This enables us to interpret the further enhancement of the MR

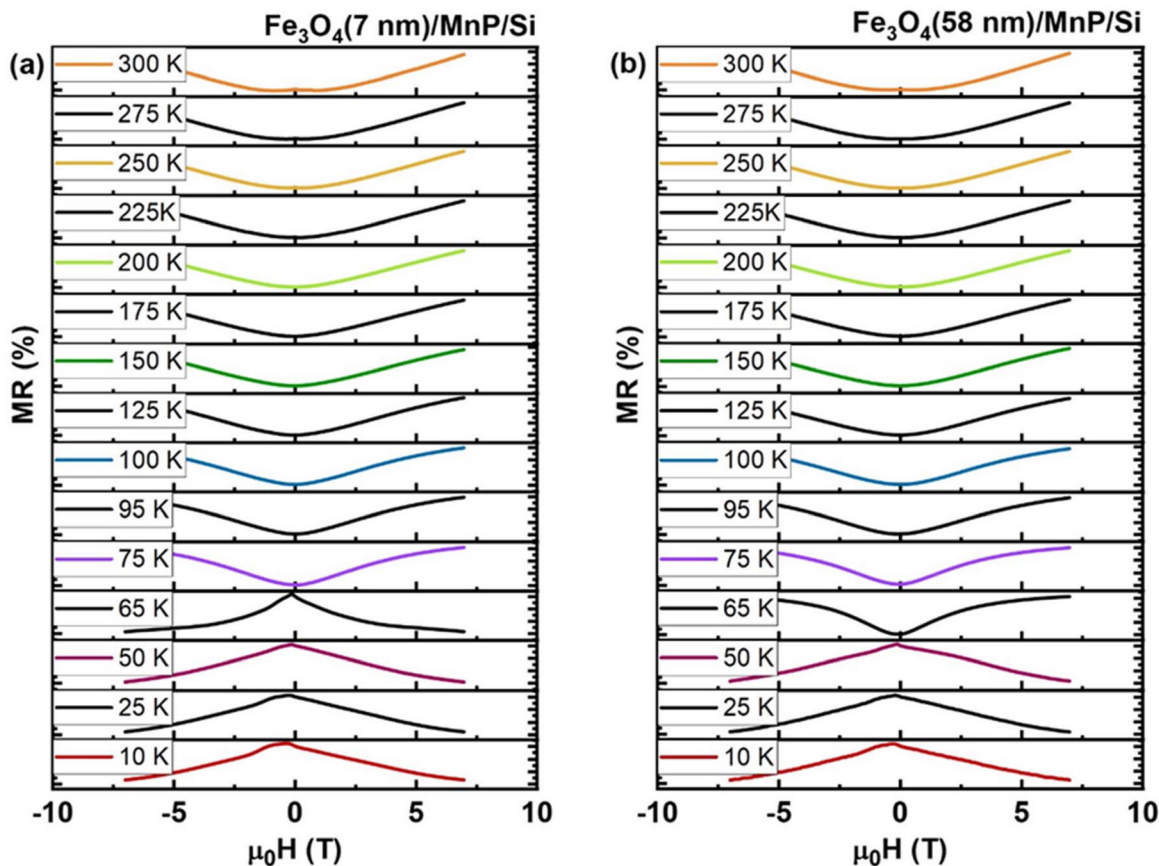


Fig. 5 In-plane (IP) magnetic field-dependent longitudinal magnetoresistance (MR) for (a) Fe_3O_4 (7 nm)/MnP/Si and (b) Fe_3O_4 (58 nm)/MnP/Si.



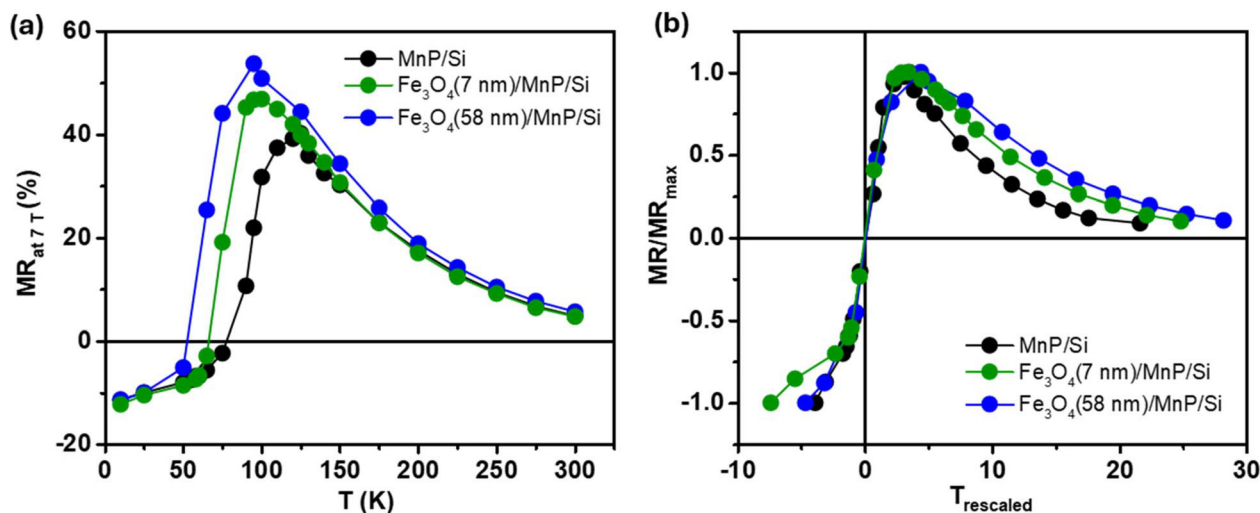


Fig. 6 (a) Temperature-dependent longitudinal magnetoresistance (MR) ratio at 7 T for MnP/Si, Fe₃O₄ (7 nm)/MnP/Si, and Fe₃O₄ (58 nm)/MnP/Si under an in-plane magnetic field and (b) the rescaled temperature-dependent normalized MR for the three films.

ratio for Fe₃O₄/MnP/Si compared to MnP/Si when the applied magnetic field is perpendicular to the plane of the film (referred to as out-of-plane (OP) MR) (Fig. S8a), in comparison with the in-plane (IP) MR case (Fig. 6a).

Furthermore, it is important to note that the peak of the MR(*T*) for the bare MnP film significantly shifts to lower temperatures in the Fe₃O₄-deposited MnP films. Increasing the thickness of the Fe₃O₄ layer further shifts the MR peak to even lower temperatures (Fig. 6). However, the overall MR(*T*) dependence retains a similar trend for all the samples. This might indicate the intrinsic transport characteristics of the MnP nanocrystalline film in the helimagnetic regime. To validate this, we have introduced a “universal curve” analytical method by rescaling the MR ratios to their maximum values and temperatures to the transition temperatures. The transition temperature (*T*_{transition}) is defined as the point where the MR at 7 T becomes zero, with a sign change from positive to negative. The respective *T*_{transition} values for MnP/Si, Fe₃O₄ (7 nm)/MnP, and Fe₃O₄ (58 nm)/MnP are indicated by color boundaries in the MR(*T*) graphs in Fig. S8(b–d). The MR ratios (%) at 7 T were normalized with respect to their maximum values, MR (%) / MR (%)^{max}, for both the positive and negative sides of MR. This normalized data was plotted against the rescaled temperature (*T*_{rescaled}), which is defined as follows.⁶⁸

$$T_{\text{rescaled}} = -\left(\frac{T - T_{\text{transition}}}{T_1 - T_{\text{transition}}}\right) \quad \text{for } T \leq T_{\text{transition}}$$

$$T_{\text{rescaled}} = \left(\frac{T - T_{\text{transition}}}{T_2 - T_{\text{transition}}}\right) \quad \text{for } T > T_{\text{transition}}$$

where *T*₁ and *T*₂ are the reference temperatures fulfilling the criteria, $\left(\frac{T - T_{\text{transition}}}{T_1 - T_{\text{transition}}}\right) = \left(\frac{T - T_{\text{transition}}}{T_2 - T_{\text{transition}}}\right) = \text{constant} = n$, with *n* set to 0.5 in this study. It can be seen in Fig. 6b that the MR/MR_{max} vs. *T*_{rescaled} curves for all three samples nearly con-

verge onto a single master curve in the temperature range corresponding to their respective FM–HM transition temperatures. This confirms that the intrinsic transport properties of the MnP nanocrystalline film in the helimagnetic regime are identical across the samples, regardless of FO₄ thickness. Since Fe₃O₄ exhibits higher resistivity at temperatures below the Verwey transition, when compared to MnP, it suggests that the overall resistivity of the Fe₃O₄/MnP/Si films is dominated by the MnP layer. This allows most of the current to tunnel through the Fe₃O₄ layer and pass through the metallic MnP at low temperatures (<120 K). As a result, the intrinsic transport properties of the underlying MnP layer in the helimagnetic state are revealed at temperatures below this temperature.⁶⁶

To elucidate the underlying mechanisms driving the low-temperature MR in our system, we first examine the correlation between MR and the magnetization. As shown in Fig. S9a for the MnP/Si film measured at 10 K, the MR maximum occurs at the coercive field. The broad single-peak MR behavior (Fig. S9a), compared with the sharp features in the first derivative of the *M*–*H* loops (*dM/dH*, inset of Fig. S9a), indicates that the magnetoresistance arises from spin-dependent scattering averaged over domain reversal rather than from abrupt magnetization switching in MnP.^{69–71} Fig. S9b shows a comparative plot of MR and $-(M/M_s)^2$ versus magnetic field at 10 K for MnP/Si. The strong agreement in the hysteresis behavior and peak positions confirms that the MR scales with the square of the magnetization, indicating that spin-dependent scattering is the dominant contribution to the observed negative MR.^{72,73} However, when similar analysis is extended to Fe₃O₄/MnP/Si, deviations from $-(M/M_s)^2$ scaling emerge. This suggests the presence of additional transport contributions such as multi-carrier conduction or interfacial scattering, which are not captured by the spin-scattering-only picture.

Finally, to get insight into the physical mechanism(s) behind the observed magnetic field dependent transport pro-



properties of the $\text{Fe}_3\text{O}_4/\text{MnP}$ bilayers, we have considered the semi empirical formula by Khosla and Fischer (KF) and its modification. The KF model explains the negative MR arising from spin-dependent scattering of carriers by localized magnetic moments,⁷⁴ but cannot fully capture the negative MR behavior in MnP/Si or Fe_3O_4 (7 nm)/ MnP/Si . To improve the fitting, we have employed the modified KF model, as expressed in eqn (1).^{36,57}

$$\text{MR}(H) = -a^2 \ln(1 + b^2 H^2) + \left(\frac{c^2 H^2}{1 + d^2 H^2} \right) \quad (1)$$

where $a = [A_1 J_{\text{ex}} D(E_F) \{S(S + 1) + M^2\}]^{1/2}$, $b = \left[1 + 4\pi^2 S^2 \left(\frac{2J_{\text{ex}} D(E_F)}{g} \right) \right]^{1/2} \left(\frac{g\mu_B}{\beta k_B T} \right)$, A_1 represents a constant for the spin dependent scattering contribution to $\text{MR}(H)$, J_{ex} is the s-d exchange integral. $D(E_F)$ is the density of state at Fermi level, S represents the spin of the localized magnetic moment, M^2 is the average of the squared magnetization, g is the Lande

g -factor, k_B is the Boltzmann constant, μ_B is the Bohr magneton and T is the temperature. The coefficients c^2 and d^2 are given by, $c^2 = \frac{\sigma_1 \sigma_2 (\mu_1 + \mu_2)^2}{(\sigma_1 + \sigma_2)^2}$ and $d^2 = \frac{(\sigma_1 \mu_2 - \sigma_2 \mu_1)^2}{(\sigma_1 + \sigma_2)^2}$, where σ_i and μ_i represent the conductivity and mobility of i^{th} carrier channel, respectively. This modified model accounts for the positive MR behavior observed in the samples at high temperatures, which is influenced by contributions from two-carrier conduction channels, while the negative MR dominates at low temperatures.

The resulting fits for Fe_3O_4 (7 nm)/ MnP/Si are shown in Fig. 7(a–d), with fitting parameters (a , b , c and d) for both Fe_3O_4 (7 nm)/ MnP/Si and MnP/Si listed in Table S2. A notable deviation from the experimental data is observed at higher fields ($>|3|$ T) at ~ 65 K for Fe_3O_4 (7 nm)/ MnP/Si (Fig. 7d) compared to MnP/Si (Fig. S10), suggesting the presence of an additional scattering mechanism, beyond the spin-dependent and impurity band scattering typically explained by the KF model. Below ~ 65 K, the MR data of the Fe_3O_4 (7 nm)/ MnP

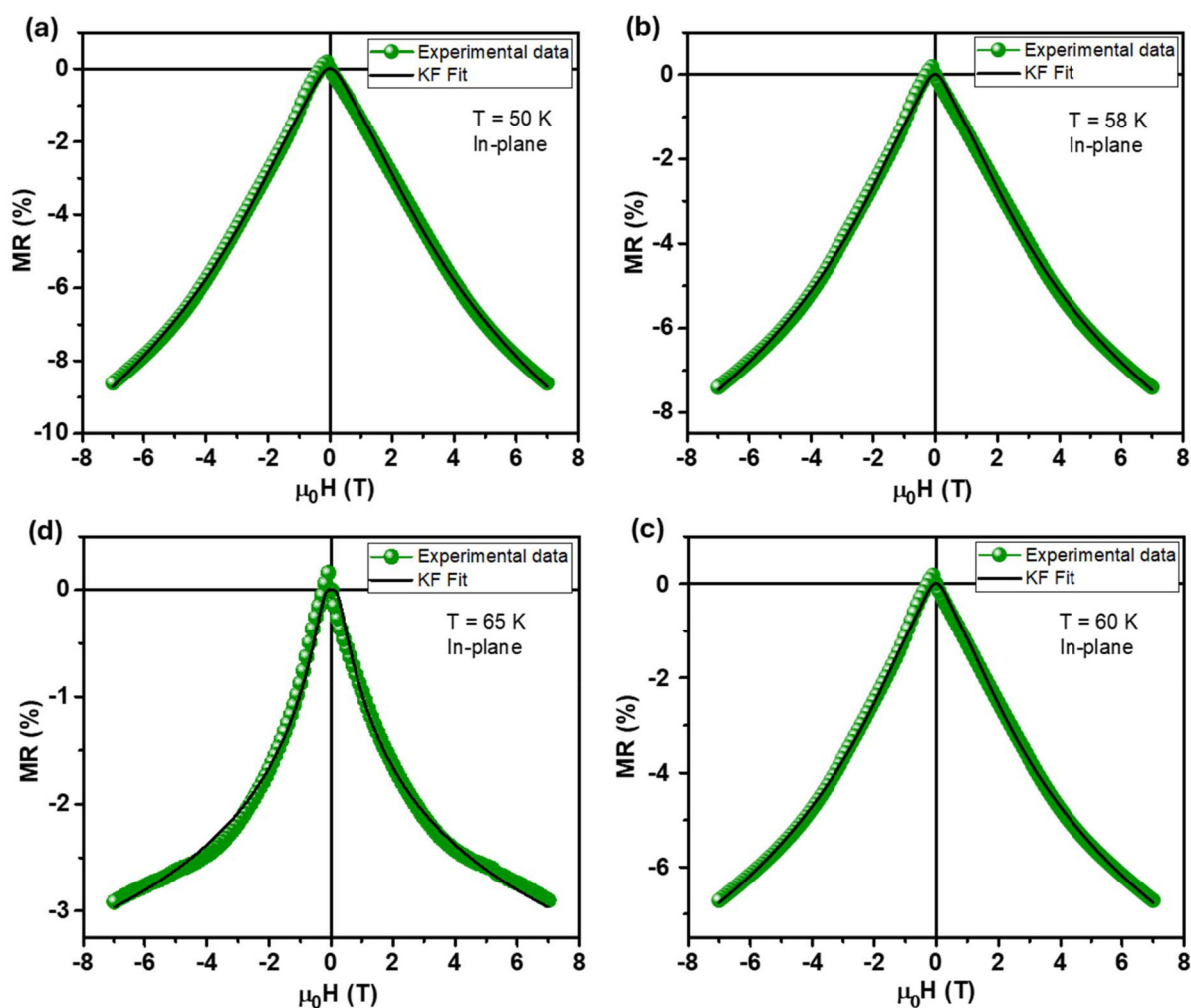


Fig. 7 In-plane (IP) magnetic field-dependent longitudinal magnetoresistance (MR) for Fe_3O_4 (7 nm)/ MnP/Si at (a) 50 K, (b) 58 K, (c) 60 K, and (d) 65 K. The solid (red) lines represent the fitting curves using the equation of Khosla and Fischer (two band) model.



bilayer, like for the bare MnP film, agrees well with the KF model, indicating that the conventional scattering mechanisms dominate at low temperatures. Interestingly, the OP MR profile of Fe₃O₄ (7 nm)/MnP/Si (Fig. S11a) at 65 K reveals additional peaks around ± 5 T, which are absent in the IP MR of Fe₃O₄ (7 nm)/MnP/Si (Fig. 7d) and the OP MR of MnP/Si (Fig. S11b). This points to the presence of complex magnetic interactions at the Fe₃O₄/MnP interface, which warrants further investigation to fully understand the underlying processes that influence the MR behavior in these bilayers.

Conclusion

In summary, this study demonstrates the successful growth of Fe₃O₄/MnP bilayers using pulsed laser deposition on MnP/Si substrates, with detailed structural, morphological, and magnetic characterizations. XRD and Raman spectroscopy confirmed the crystalline phases and proper growth of Fe₃O₄ and MnP, with notable differences in lattice parameters and surface morphology between Fe₃O₄ films grown on MnP and Si substrates. The Fe₃O₄/MnP interface exhibited distinct magnetic properties compared to Fe₃O₄/Si, likely due to strain and interfacial effects, as reflected in the surface roughness, grain size distribution, and magnetic domain structure. Magnetic force microscopy revealed stronger magnetic interactions and higher stray fields in the Fe₃O₄/MnP bilayers compared to Fe₃O₄/Si films, with variations in $\Delta\phi_{\text{RMS}}$ indicating substrate-dependent growth mechanisms. Magnetometry measurements indicate that the Fe₃O₄ layer significantly influences the magnetic properties of the MnP substrate, especially at lower temperatures. A shift in the ferromagnetic to helimagnetic transition of MnP from ~ 102 K to ~ 90 K in the Fe₃O₄/MnP bilayers was observed, attributed to strain-induced effects caused by the structural changes of Fe₃O₄ during its Verwey transition. The presence of the 7 nm-thick Fe₃O₄ layer enhances the Fe₃O₄/MnP interface coupling, resulting in the enhanced coercivity and saturation magnetization in the Fe₃O₄ (7 nm)/MnP bilayer compared to the bare MnP film. This can be attributed to the dual magnetic proximity effect and the impact of the structural transformation during the Verwey transition. In contrast, the strong soft ferromagnetic signal of the 58 nm-thick Fe₃O₄ layer dominates the overall magnetism of the Fe₃O₄ (58 nm)/MnP bilayer.

Furthermore, the temperature-dependent resistivity of Fe₃O₄/MnP bilayers revealed modifications in resistivity profiles due to the bilayer structure. The resistivity peak of MnP shifted to lower temperatures in the bilayers, indicating interfacial effects and the influence of Fe₃O₄ Verwey transition on the electronic transport properties of MnP. The MR magnitude showed significant enhancement with the Fe₃O₄ deposition, with thicker Fe₃O₄ layers (58 nm) yielding a greater MR effect compared to thinner layers (7 nm), further supporting the impact of interfacial coupling on the transport and magnetotransport properties. Overall, the results highlight the critical role of substrate and interfacial effects in determining the

magnetic and electronic properties of Fe₃O₄/MnP bilayers, providing valuable insights for spintronic device applications. While our transport measurements suggest that Fe₃O₄ deposition modifies the low-temperature electronic response of MnP, direct studies of the valence band structure at the Fe₃O₄/MnP interface are not yet available. Future experimental (*e.g.*, ARPES, XPS) and theoretical work could provide deeper insight into the electronic reconstruction at this interface and its role in the observed transport phenomena.

Experimental methods

This study utilized MnP nanostructured thin films grown on Si substrates (MnP/Si) at a substrate temperature of 500 °C using molecular beam epitaxy. The growth of MnP has been documented elsewhere.³⁴ The MnP films used in this work had an average grain size of ~ 86 nm (film thickness ~ 100 nm), which corresponds to the larger-grained films that in our previous study exhibited a maximum MR of $\sim 40\%$, as opposed to the smaller-grained (~ 39 nm) films that showed $\sim 90\%$ MR.³⁶ These larger-grained MnP films are chosen here as the baseline to evaluate the enhancement achievable by Fe₃O₄ overlayer deposition.

The MnP film was chemically cleaned (30 min in acetone, 30 min in methanol and 30 min in DI water with sonication) prior to characterizations, magnetic and transport measurements. Fe₃O₄ films with thicknesses of 7 nm and 58 nm were deposited on chemically cleaned MnP (Fe₃O₄/MnP/Si), while Fe₃O₄ films with thicknesses of 10 nm and 62 nm were grown directly on Si (Fe₃O₄/Si) using the pulsed laser deposition (PLD) technique. For the ablation, a focused KrF excimer laser (248 nm) and a commercially available Fe₃O₄ target were employed. The substrate temperatures for both Si and MnP during growth were maintained at 350 °C, with the chamber pressure held at 2×10^{-5} – 2.2×10^{-5} Torr. The laser fluence was set at 2 J cm⁻², and the target-to-substrate distance was kept at 6 cm during deposition, with a repetition rate of 6 Hz. After deposition, the films were cooled to room temperature at a rate of 2 °C min⁻¹ under the same chamber conditions as during growth. The presence of the Fe₃O₄ phase in the films was verified using Raman spectroscopy. Crystallinity of the MnP/Si, Fe₃O₄/MnP/Si bilayers, and Fe₃O₄/Si was assessed through X-ray diffraction (XRD, RIGAKU SmartLab). The presence of elements (Fe, O, Mn and P) in Fe₃O₄/MnP bilayers were confirmed using energy dispersive spectroscopy (EDS, Hitachi SU-70). Morphological analysis was performed using scanning electron microscopy (SEM, JEOL JSM-6390LV), while topography and root-mean-square (RMS) roughness were determined by atomic force microscopy (AFM, Hitachi AFM5300E) at room temperature and under vacuum. Magnetic domain images of the MnP and Fe₃O₄/MnP bilayers were captured using magnetic force microscopy (MFM) under similar conditions. Magnetometry and resistivity measurements were conducted using vibrating sample magnetometry and DC resistivity options of a Quantum Design Physical Property Measurement



System. Temperature-dependent magnetization ($M-T$) was measured under three protocols: zero-field cooling (ZFC), field-cooled cooling (FCC), and field-cooled warming (FCW) across a temperature range of 10 K to 350 K under a 0.1 T magnetic field. Field-dependent magnetization ($M-H$) was assessed under magnetic fields of ± 5 T within the same temperature range for the MnP and $\text{Fe}_3\text{O}_4/\text{MnP}$ bilayers. Finally, temperature-dependent resistivity ($\rho(T)$) and magnetic field-dependent longitudinal and transverse resistivity (MR) measurements were performed using a standard four-probe configuration across a temperature range of 10 K to 350 K and under magnetic fields of +7 T to -7 T (unipolar sweep). In this configuration, the magnetic field (H) were applied either along the plane of the film (IP) or perpendicular to the plane of the film (OP).

Author contributions

Conceptualization, investigation, methodology, writing – original draft, and writing – review and editing, N. W. Y. A. Y. M.; investigation and writing – review and editing, D. D., and A. C.; investigation and writing – review, Y. M. W., and E. O.; resources, writing – review and editing, A. T. D., H. R. G., X. J., and S. W.; conceptualization, funding acquisition, supervision, and writing – review and editing, M. H. P.

Conflicts of interest

The authors declare no conflicts of interest.

Data availability

The raw data required to reproduce the findings cannot be shared at this time as the data also forms part of an ongoing study.

Supplementary information (SI) is available. See DOI: <https://doi.org/10.1039/d5nr02716k>.

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