

Heterointerface-enhanced magnetic properties of $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$ composite fibers

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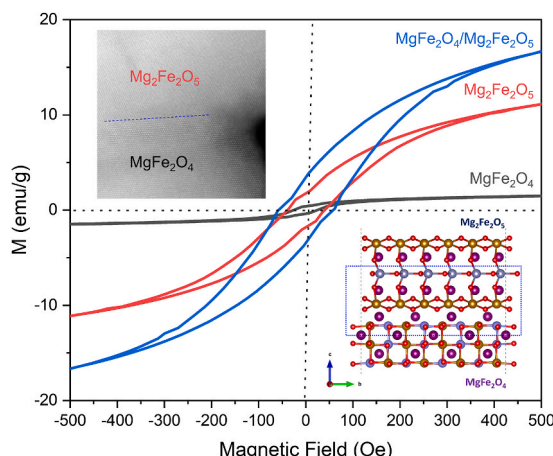
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HIGHLIGHTS

- Heterointerfaces between MgFe_2O_4 and $\text{Mg}_2\text{Fe}_2\text{O}_5$ are constructed in composite fibers via electrospinning and annealing.
- The composite fibers show improved saturation magnetization and magnetic anisotropy compared to single-phase counterparts.
- Interfacial electronic coupling suppresses spin-down states of Fe atoms, boosting the net magnetic moment.

GRAPHICAL ABSTRACT



ABSTRACT

Magnesium-based ferrites demonstrate significant potential for applications in electromagnetic and spintronic devices, but their practical utilization is often restricted by their relatively low magnetization. Heterointerface engineering provides a promising way to overcome this challenge by tuning magnetic interactions at the boundaries between different phases. In this work, $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$ composite fibers are synthesized via electrospinning followed by a high-temperature annealing in air. Microstructural characterization reveals the formation of abundant heterointerfaces in the composite fibers with well-defined orientation relationship. Compared with pure MgFe_2O_4 and $\text{Mg}_2\text{Fe}_2\text{O}_5$ fibers, the composite fibers exhibit significantly enhanced saturation magnetization and magnetic anisotropy, demonstrating a positive effect of heterointerface engineering on magnetic performance. Combining experimental results with first-principles calculations, the underlying mechanism for the magnetization enhancement is elucidated. It reveals that the interfacial interactions between MgFe_2O_4 and $\text{Mg}_2\text{Fe}_2\text{O}_5$ suppress the spin-down states of Fe atoms, thereby boosting the net magnetic moment of the composite. This work reveals the synergistic effects of interfacial electronic coupling and spin polarization, providing a practical path for the design of high-magnetization soft ferrites suitable for miniaturized microwave devices and high-density magnetic storage.

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1. Introduction

Soft magnetic ferrites are indispensable components of modern electronic devices. Driven by the rapid advancement of 5G communications, electric vehicles, and an ongoing trend towards device miniaturization, the demand for these materials is experiencing unprecedented growth [1–8]. As a class of ferrimagnetic oxides primarily composed of Fe_2O_3 , soft magnetic ferrites are characterized by high electrical resistivity, low coercivity, and high magnetic permeability, making them ideal for high-frequency applications such as transformers, inductors, and noise filters [9–14]. Although these ferrites are easily magnetized and demagnetized, their relatively low saturation magnetization (M_s) remains a primary bottleneck, restricting their performance in high-density power and sensing applications [15–17].

Among various ferrites, magnesium ferrite (MgFe_2O_4), an n-type semiconductor with a partially inverse spinel structure, has attracted significant attention due to its excellent chemical stability, low cost, and unique biocompatibility [18–25]. These attributes set MgFe_2O_4 apart from the transition-metal ferrites, extending its usage from traditional electronics to biomedical realms, such as magnetic resonance imaging contrast agents and targeted drug delivery [26–30]. However, the M_s of cubic MgFe_2O_4 is inherently constrained by its cation distribution, which limits the net magnetic moment and hinders its practical application in high-performance devices. Therefore, developing effective strategies to enhance the magnetization of MgFe_2O_4 has attracted extensive research attention in recent years. In the Mg-Fe-O system, orthorhombic $\text{Mg}_2\text{Fe}_2\text{O}_5$ also exhibits soft magnetic characteristics [31, 32]. Compared with cubic MgFe_2O_4 , the orthorhombic $\text{Mg}_2\text{Fe}_2\text{O}_5$ features lower crystallographic symmetry, which could potentially give rise to a higher magnetic anisotropy. Nevertheless, systematic comparative investigation is still scarce on the magnetic behaviors of MgFe_2O_4 and $\text{Mg}_2\text{Fe}_2\text{O}_5$, and the magnetic performance modification through coupling these two different phases remains underexplored.

In recent years, research work on MgFe_2O_4 and $\text{Mg}_2\text{Fe}_2\text{O}_5$ has predominantly focused on photocatalytic, gas-sensing, and microwave absorption properties [33–39]. For instance, heterostructured systems such as $\text{MgFe}_2\text{O}_4/\text{CuO}$ and $\text{Mg}_2\text{Fe}_2\text{O}_5$ -decorated CaFe_2O_4 have demonstrated significantly enhanced charge transfer and catalytic efficiency compared to their single-phase counterparts [25,33]. These findings suggest that constructing heterointerfaces can fundamentally modify the interfacial electronic environment and band structures. In terms of magnetism, some studies, *i.e.*, $\text{MgFe}_2\text{O}_4/\text{MnO}_2$ nanohybrids [40], have achieved an altered magnetic hysteresis behavior upon composite formation. However, the fundamental mechanism regarding how heterointerfaces between two magnesium ferrites, *i.e.*, MgFe_2O_4 and $\text{Mg}_2\text{Fe}_2\text{O}_5$, influence the magnetic anisotropy and the net magnetic moment remains unclear.

In this work, $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$ composite fibers were fabricated *via* electrospinning followed by a high-temperature annealing. By precisely tailoring the Mg/Fe molar ratio, many heterointerfaces between MgFe_2O_4 and $\text{Mg}_2\text{Fe}_2\text{O}_5$ have been achieved, with well-defined orientation relationship. It reveals that the composite fibers exhibit significantly enhanced magnetic anisotropy constant and saturation magnetization compared to both MgFe_2O_4 and $\text{Mg}_2\text{Fe}_2\text{O}_5$. The underlying mechanism for the enhanced magnetization has been elucidated by combining magnetic hysteresis loop measurements with first-principles calculations.

Table 1
Raw material ratios for different products.

Raw materials			Products
Mg(acac) ₂ (g)	Fe(acac) ₃ (g)	Molar ratio	
0.22	0.70	1:2	MgFe_2O_4
0.22	0.35	1:1	$\text{Mg}_2\text{Fe}_2\text{O}_5$
0.22	0.52	1:1.5	$\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$

2. Experimental

2.1. Materials

Magnesium acetylacetonate [$\text{Mg}(\text{acac})_2$] and iron acetylacetonate [$\text{Fe}(\text{acac})_3$] powders, polyvinylpyrrolidone (PVP, molecular weight: 1,300,000), were purchased from Sinopharm Chemical Reagent Company Limited. All chemicals were of analytical grade and used as received without further purification.

2.2. Preparation of fibers

MgFe_2O_4 , $\text{Mg}_2\text{Fe}_2\text{O}_5$, and $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$ fibers were fabricated *via* the electrospinning technique. $\text{Mg}(\text{acac})_2$ and $\text{Fe}(\text{acac})_3$ powders, in varying molar ratios (detailed in Table 1), were precisely weighed and placed into a reagent bottle. Subsequently, 0.66 g of PVP was added into the bottle, followed by sequential addition of 3 mL of anhydrous ethanol and 3 mL of glacial acetic acid. The resulting solution was then subjected to a continuous stirring for 6 h to ensure formation of a homogeneous and viscous solution. Next, 3–5 mL of this solution was loaded into a syringe for electrospinning.

The electrospinning parameters were optimized as follows [41–43]: a spinning voltage of 16 kV was applied between the needle and the collector with a distance of 15 cm; the injection rate was 0.5 mL/h; the collector of the fibers consisted of a steel plate wrapped with oil paper rotating at 500 rpm; the temperature was 25 °C; the humidity was ~45%. After the electrospinning process, the resulting fiber membrane was carefully removed from the collector drum and dried at 60 °C for 2 h to remove any residual solvents. After drying, the fiber membranes were then annealed at 900 °C for 12 h at a heating rate of 5 °C/min in air. The final products were obtained after the annealing process. The obtained fiber membranes with Mg:Fe ratios of 1:1, 1:2 and 1:1.5 were named as MF-11, MF-12 and MF-23, respectively.

2.3. Materials characterization

The crystal structures of three products were analyzed by an X-ray diffractometer (XRD, Bruker D8 Advance) with $\text{Cu-K}\alpha$ radiation ($\lambda = 1.5406 \text{ \AA}$). The morphology of the products was examined using a field-emission SEM (FE-SEM, Sigma 500) operating at 15 kV. X-ray photoelectron spectroscopy (XPS) spectra were acquired using a Thermo Scientific K-Alpha⁺ XPS spectrometer equipped with an Al $\text{K}\alpha$ X-ray source. Bright-field (BF) images, high-resolution transmission electron microscopy (HRTEM) images and selected-area electron diffraction (SAED) patterns were obtained using a field-emission transmission electron microscope (TEM, JEOL JEM2100F) operating at 200 kV. The elemental mapping of the fibers was carried out using energy dispersive X-ray spectrometer (EDS, Oxford INCAx-sight 6427). The high-angle annular dark field (HAADF) imaging was performed using a probe-corrected Thermo Scientific Spectra 300 (S)TEM with an accelerating voltage of 300 kV. The magnetization *versus* magnetic field hysteresis loops were measured by a vibrating sample magnetometer (VSM) equipped on a physical properties measurement system (PPMS, Quantum Design) at room temperature.

2.4. Computational methodology

Spin-polarized electronic structure calculations for MgFe_2O_4 , $\text{Mg}_2\text{Fe}_2\text{O}_5$ and $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$ interface were performed using the plane-wave basis set method, as implemented in the Vienna *ab-initio* Simulation Package (VASP) [44]. These calculations were conducted with initial lattice parameters of $a = 8.38 \text{ \AA}$ for MgFe_2O_4 and $a = 13.30 \text{ \AA}$, $b = 5.19 \text{ \AA}$, $c = 5.17 \text{ \AA}$ for $\text{Mg}_2\text{Fe}_2\text{O}_5$, respectively. The projector augmented wave pseudopotential (PAW) was employed to describe the interactions between ions and core electrons. The exchange and correlation effects were described using the Perdew-Burke-Ernzerhof (PBE)

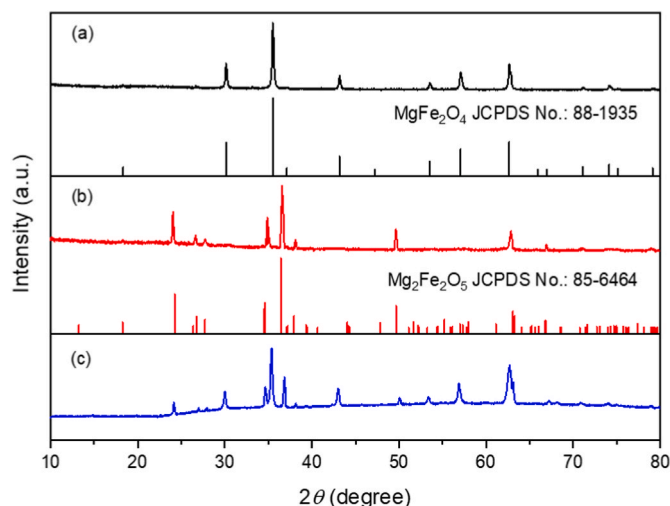


Fig. 1. XRD patterns obtained from MF-12 (a), MF-11 (b), and MF-23 (c).

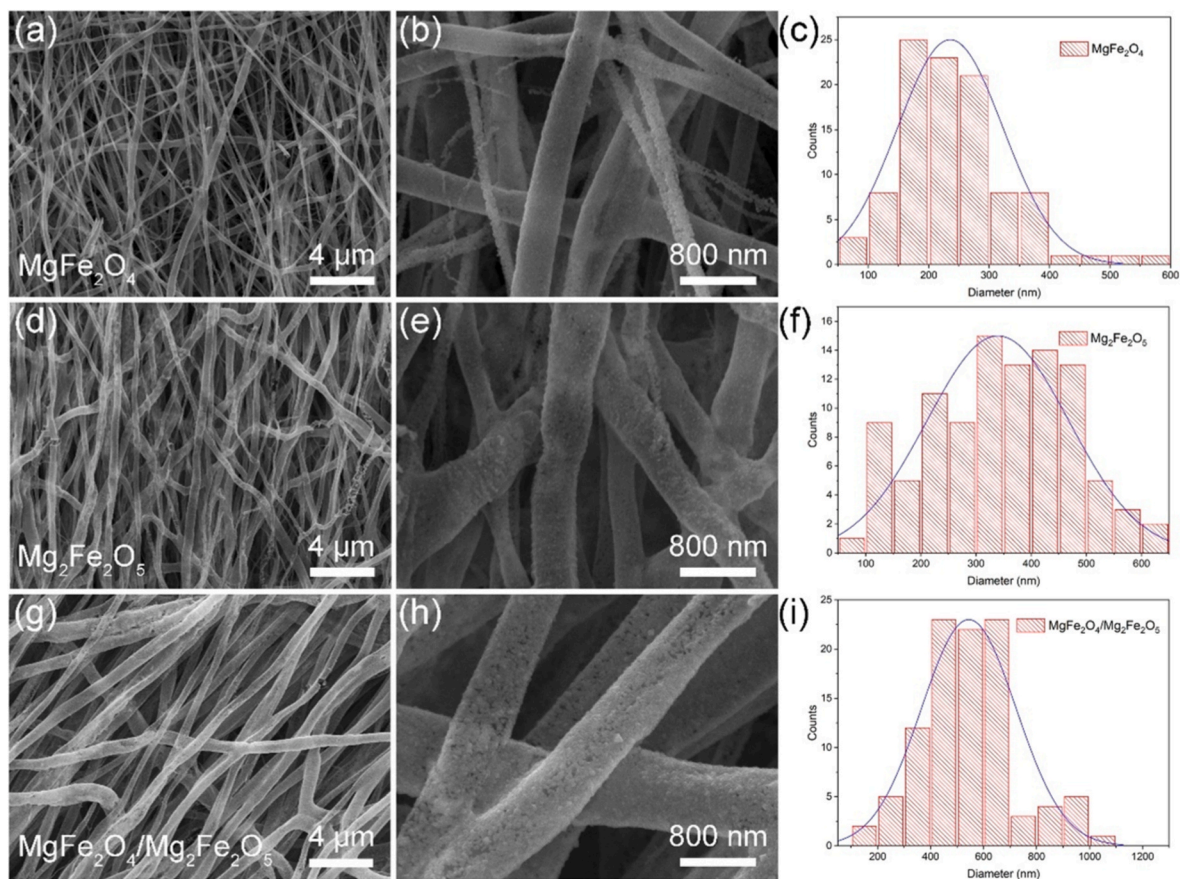


Fig. 2. SEM images and diameter distributions of MgFe_2O_4 (a, b, c), $\text{Mg}_2\text{Fe}_2\text{O}_5$ (d, e, f) and $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$ (g, h, i) submicron fibers.

within the framework of generalized gradient approximation (GGA) [45]. The cutoff energy for the truncation of plane-wave basis was set to be 520 eV, and the accuracy of the electron self-consistent field was set to be 10^{-6} eV. A Brillouin zone k -point sampling with a resolution of $2\pi \times 0.025 \text{ \AA}^{-1}$ was used. The convergence criterion is 0.01 eV/ $\text{\AA}$ for the forces acting on the atoms. The self-consistent spin-polarized calculations were performed for collinear magnetism. The Fe d -projected partial density of states (PDOS) were calculated to analyze the magnetic properties. The calculation process for PDOS of Fe at different sites in

MgFe_2O_4 , $\text{Mg}_2\text{Fe}_2\text{O}_5$ unit cells and $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$ interface can be found in Supporting Information.

3. Results and discussion

3.1. Morphology and crystal structure of magnesium-based ferrite fibers

To analyze the crystal structure of the as-prepared products, XRD analysis was performed, as presented in Fig. 1. As shown in the XRD

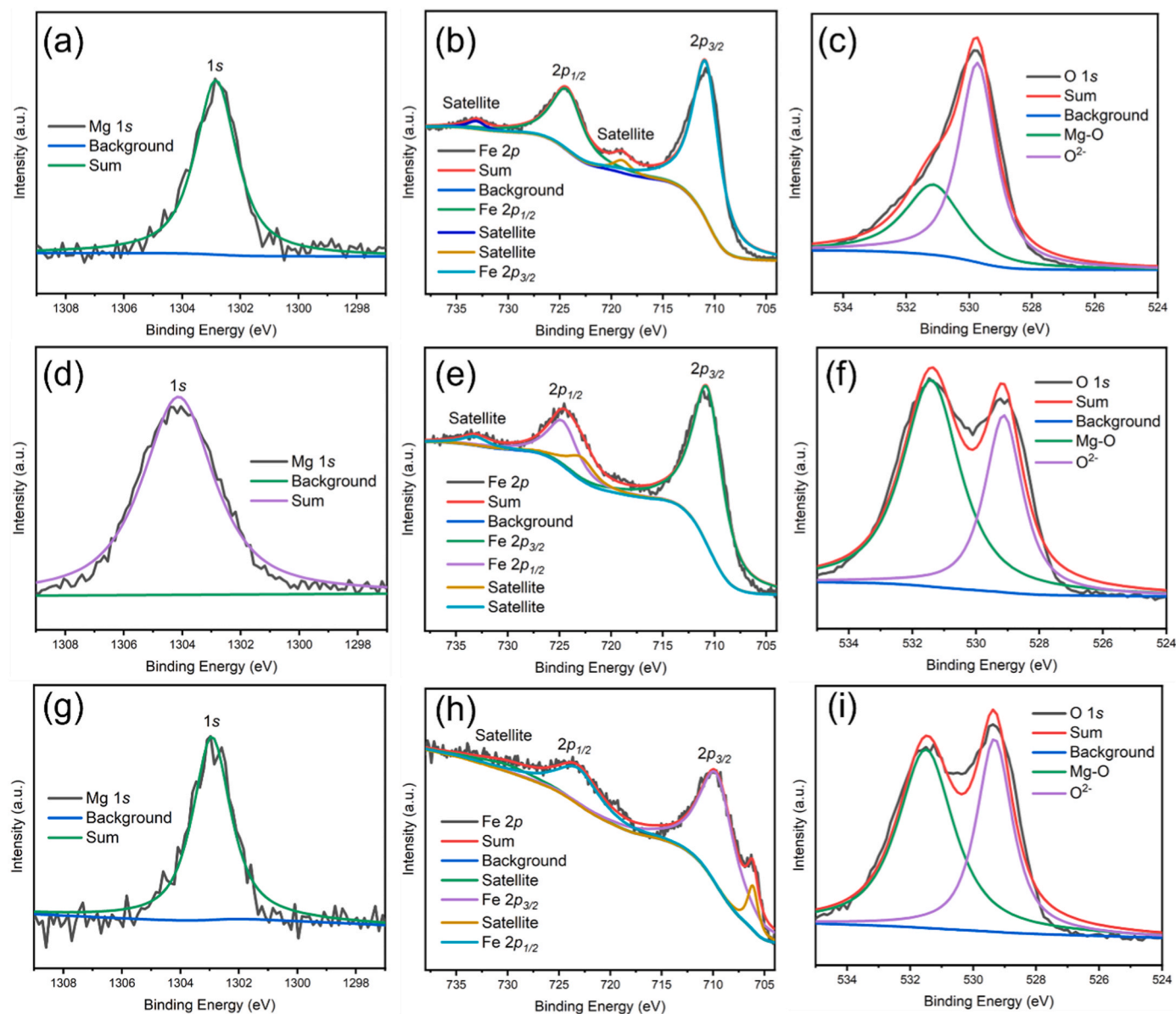


Fig. 3. XPS spectra of Mg, Fe, and O in MgFe_2O_4 (a, b, c), $\text{Mg}_2\text{Fe}_2\text{O}_5$ (d, e, f), and $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$ (g, h, i) submicron fibers.

patterns, when the molar ratio of $\text{Mg}(\text{acac})_2$ and $\text{Fe}(\text{acac})_3$ in the precursor is 1:2, all diffraction peaks can be indexed using the cubic MgFe_2O_4 (JCPDS No.: 88-1935; $a = 8.38 \text{ \AA}$). The peaks at 30.1° , 35.5° , 43.1° , 53.5° , 57.1° , and 62.7° correspond to (220), (311), (400), (422), (511), and (440) planes of MgFe_2O_4 , respectively. Upon adjusting the molar ratio to 1:1, the diffraction peaks match the standard pattern of orthorhombic $\text{Mg}_2\text{Fe}_2\text{O}_5$ (JCPDS No.: 85-6464; $a = 13.30 \text{ \AA}$, $b = 5.19 \text{ \AA}$, $c = 5.17 \text{ \AA}$). The peaks at 24.2° , 34.5° , 36.4° , 49.7° , and 63.1° are assigned to (011), (002), (411), (022), and (431) planes of $\text{Mg}_2\text{Fe}_2\text{O}_5$, respectively. At an intermediate molar ratio of 1:1.5, the XRD pattern exhibits characteristic diffraction peaks from both MgFe_2O_4 and $\text{Mg}_2\text{Fe}_2\text{O}_5$, confirming the formation of a composite. Notably, the diffraction peaks of MgFe_2O_4 display higher intensity than those of $\text{Mg}_2\text{Fe}_2\text{O}_5$. Quantitative analysis reveals that the molar ratio of MgFe_2O_4 and $\text{Mg}_2\text{Fe}_2\text{O}_5$ is approximately 2:1. This corresponds to an overall Mg:Fe atomic ratio of 1:1.5 in the composite, consistent with the precursor stoichiometry used in the synthetic process. Furthermore, all three products exhibit excellent crystallinity, as evidenced by the sharp and well-defined diffraction peaks.

To investigate the morphology and size distribution of three ferrites,

MgFe_2O_4 , $\text{Mg}_2\text{Fe}_2\text{O}_5$, and $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$, extensive SEM examinations were conducted, as shown in Fig. 2. All three products exhibit a fibrous morphology with shared characteristics: rough surfaces formed by dense aggregation and interconnection of numerous small crystallites. The morphological and dimensional comparisons are listed as follows. For the MgFe_2O_4 submicron fibers, low- and high-magnification SEM images in Fig. 2(a) and (b) reveal relatively straight fiber morphology, albeit with noticeable variation in diameter uniformity. Statistical size analysis from Fig. 2(c) indicates that most fibers have diameters within the 150–300 nm range, yielding an average diameter of $\sim 200 \text{ nm}$. High-magnification imaging further discloses a rough surface composed of extremely fine and densely packed crystallites. The $\text{Mg}_2\text{Fe}_2\text{O}_5$ submicron fibers, as shown in Fig. 2(d) and (e), display a slightly larger average diameter of about 400 nm and exhibit pronounced curvature or bending. The fiber surface remains rough with tight interconnection of ultrafine crystallites, mirroring the microstructural features observed in MgFe_2O_4 fibers. The $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$ composite submicron fibers, as shown in Fig. 2(g) and (h), demonstrate superior macroscopic alignment and uniformity, with no observable fractures and more homogeneous size distribution compared to the

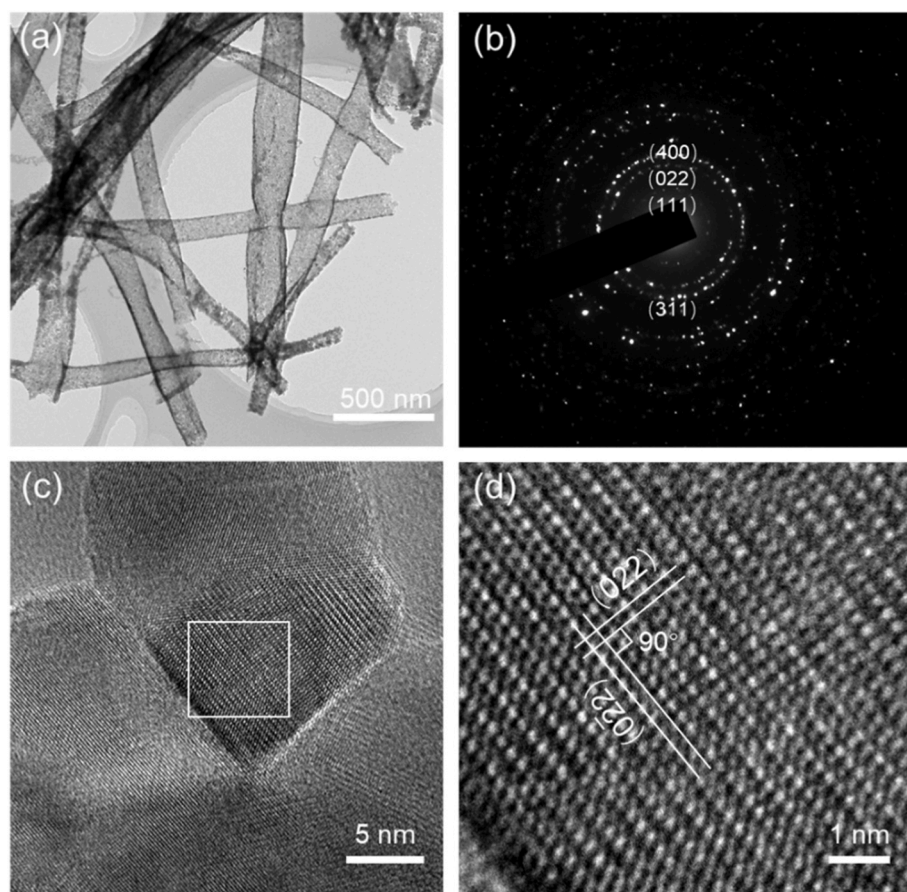


Fig. 4. TEM images of MgFe_2O_4 submicron fibers. (a) BF image, (b) SAED pattern of the fibers in (a), (c) HRTEM image of a localized region of a single grain, (d) Enlarged view of the rectangular region in (c).

single-phase counterparts. The composite fiber diameters increase markedly, ranging from 300 to 700 nm, with an average of 550 nm as determined from the statistical analysis in Fig. 2(i). High-magnification SEM reveals that the surface roughness and granular microstructure are analogous to those of the single-phase fibers, originating from accumulation of abundant minute crystallites. These observations indicate that phase composition, controlled by adjusting the precursor stoichiometry, profoundly influences the fiber diameter, curvature and uniformity while preserving the rough characteristic and polycrystalline fibrous architecture typical of the ferrites fabricated through electrospinning and subsequent annealing [46–51].

To investigate the surface elemental composition and chemical valence states of the three ferrites, XPS measurements were performed, as presented in Fig. 3. Fig. 3(a–c) illustrate the Mg 1s, Fe 2p, and O 1s spectra of the MgFe_2O_4 fibers. In Fig. 3(a), the peak at 1303.1 eV corresponds to the Mg 1s orbit of Mg^{2+} , confirming the presence of Mg^{2+} in MgFe_2O_4 [52]. The Fe 2p spectrum in Fig. 3(b) shows characteristic peaks at 710.7 eV (Fe $2p_{3/2}$) and 724.3 eV (Fe $2p_{1/2}$), along with satellite peaks at approximately 718 eV and 733 eV, indicating the presence of Fe^{3+} [53]. The O 1s spectrum in Fig. 3(c) exhibits two distinct components at 529.9 eV and 531.5 eV, corresponding to lattice oxygen (O^{2-} in the crystal structure) and surface-adsorbed oxygen species, respectively [46]. Fig. 3(d–f) present the corresponding Mg 1s, Fe 2p, and O 1s spectra obtained from $\text{Mg}_2\text{Fe}_2\text{O}_5$ submicron fibers. The Mg 1s peak at 1303.1 eV confirms the presence of Mg^{2+} [54]. The Fe 2p spectrum in Fig. 3(e) is similar to that of MgFe_2O_4 , with Fe $2p_{3/2}$ at 710.7 eV, Fe $2p_{1/2}$ at 724.3 eV, and satellite features at ~718 eV and 733 eV, verifying the presence of Fe^{3+} . The O 1s spectrum in Fig. 3(f) also resolves lattice oxygen at 529.9 eV and adsorbed oxygen at 531.5 eV. Notably, the relative intensity of the adsorbed oxygen peak is significantly

enhanced compared to MgFe_2O_4 , which suggests a higher concentration of oxygen vacancies or a more active surface area in the $\text{Mg}_2\text{Fe}_2\text{O}_5$. Fig. 3 (g–i) show the typical high-resolution spectra of Mg 2p, Fe 2p, and O 1s for the $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$ composite submicron fibers. The elemental composition of the composite is confirmed to be Mg, Fe, and O. The binding energies and peak shapes indicate that the valence states in the composite remain consistent with those in individual components (Mg^{2+} , Fe^{3+} , and O^{2-}). Notably, an additional peak at ~706 eV appears in the Fe 2p spectrum of the $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$ composite fibers, which is absent in the spectra of both MgFe_2O_4 and $\text{Mg}_2\text{Fe}_2\text{O}_5$ fibers. Such a change is likely associated with the formation of $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$ heterointerfaces, where interfacial interactions alter the local electronic environment of Fe atoms near the heterointerfaces. A similar phenomenon has also been observed in $\text{CaFe}_2\text{O}_4/\text{Ca}_2\text{Fe}_2\text{O}_5$ composite fibers [55], which is ascribed to the electronic structure modulation of partial Fe atoms in the composite fibers.

3.2. Microstructural characterization of magnesium-based ferrite fibers

To gain deeper insights into the microstructural features of MgFe_2O_4 , $\text{Mg}_2\text{Fe}_2\text{O}_5$, and $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$ submicron fibers, extensive TEM examinations were conducted. Fig. 4 presents the TEM images for the MgFe_2O_4 submicron fibers. The BF TEM image in Fig. 4(a) reveals that the fiber is composed of numerous ultrafine nanocrystals. The corresponding SAED pattern in Fig. 4(b) displays polycrystalline diffraction rings, which can be indexed, from inner to outer, as the (111), (220), (311), and (400) planes of cubic MgFe_2O_4 . HRTEM image in Fig. 4(c) shows several nanocrystals with varying orientations and sizes ranging from 5 to 20 nm. The lattice fringes within individual grains are continuous and sharp, indicating good crystallinity. As shown in Fig. 4

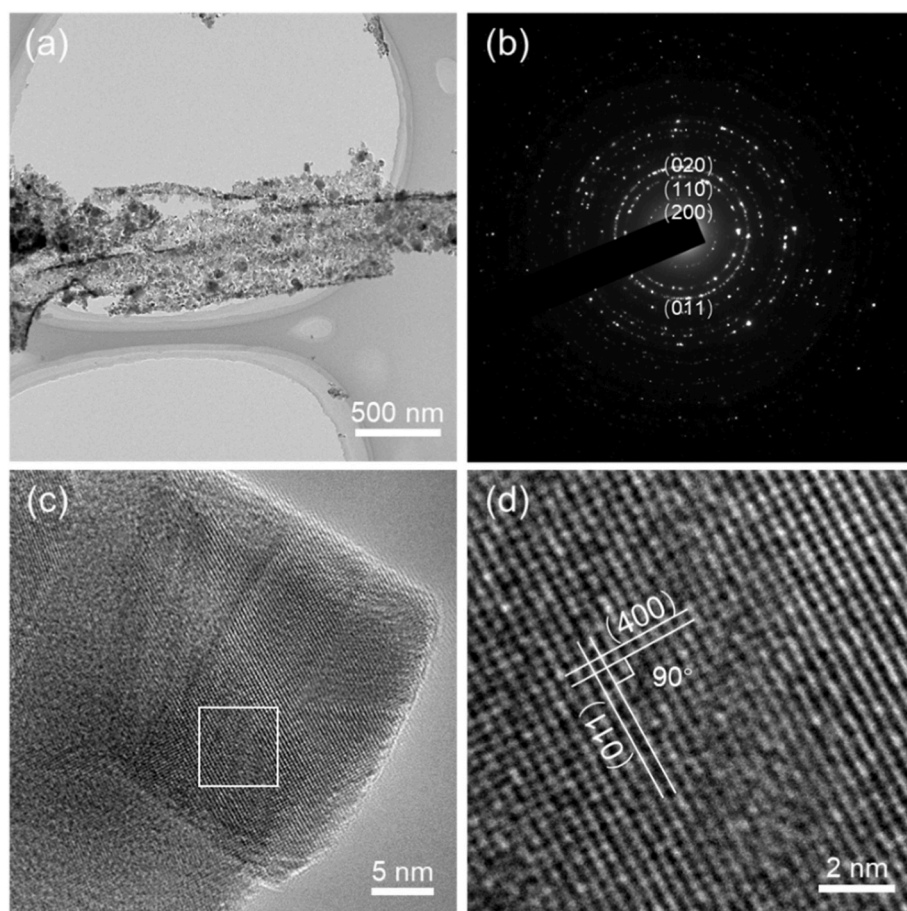


Fig. 5. TEM images of $\text{Mg}_2\text{Fe}_2\text{O}_5$ submicron fibers. (a) BF TEM image, (b) Corresponding SAED pattern, (c) HRTEM image of a localized region of a single grain, (d) Enlarged image of the region enclosed by a rectangle in (c).

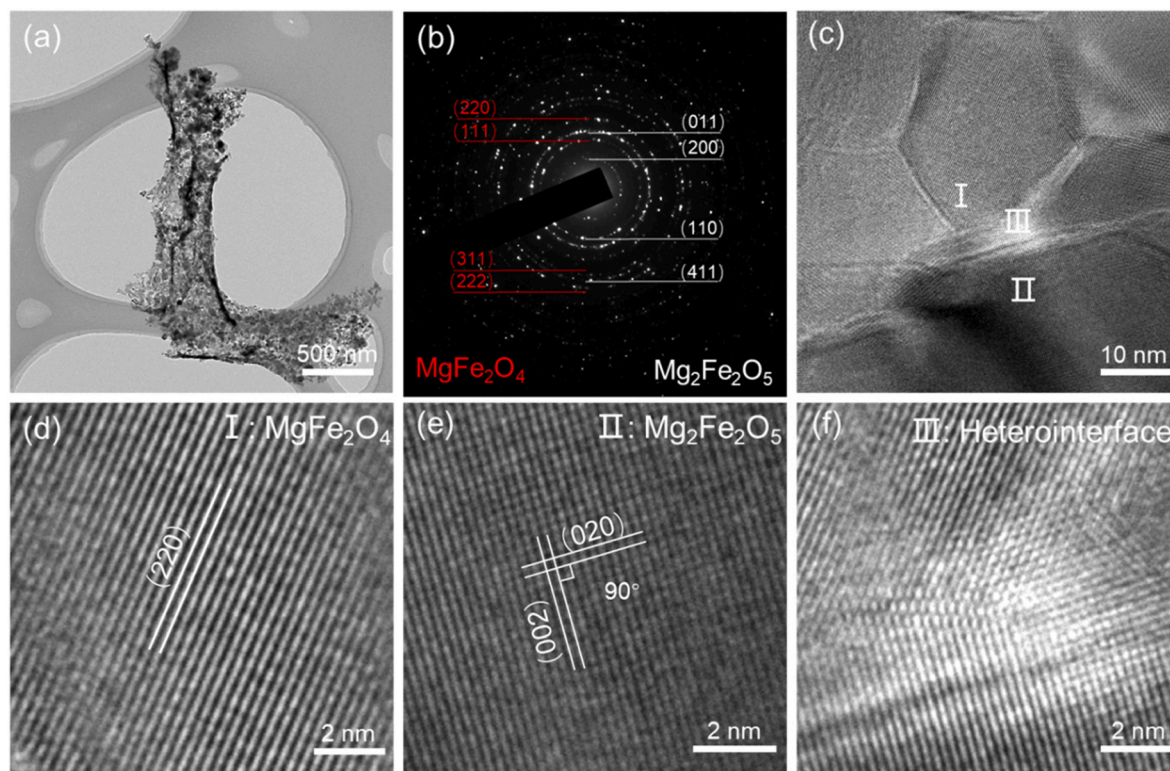


Fig. 6. TEM images of $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$ submicron composite fibers. (a) BF TEM image, (b) SAED patterns obtained from the fiber in (a), (c) HRTEM image of nanograins of fiber in (a), The enlarged HRTEM images obtained from grain I (d), grain II (e), and grain III (f).

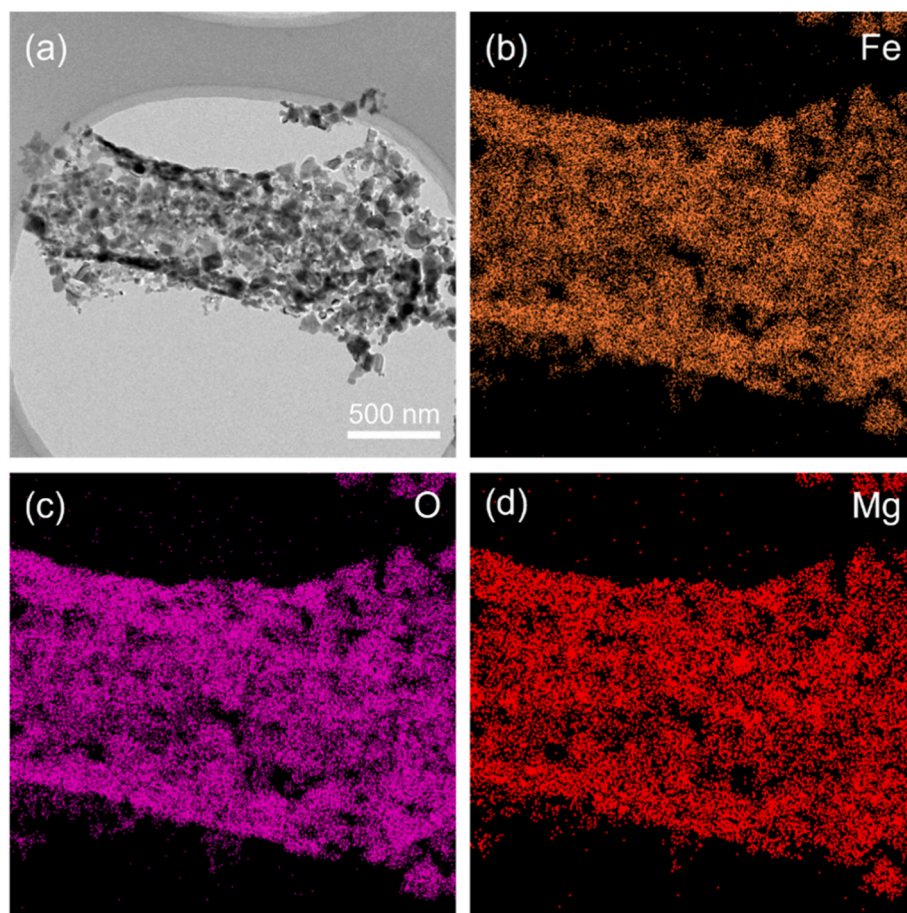


Fig. 7. (a) TEM image of $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$ composite fibers; EDS elemental mapping: (b) Fe, (c) O, and (d) Mg.

(d), the enlarged HRTEM image of the region enclosed by a white square in Fig. 4(c) reveals an interplanar spacing of 2.96 Å and an interplanar angle of 90°, which can be indexed as the (2 2̄ 0) and (022) planes of cubic MgFe_2O_4 . These observations confirm that the MgFe_2O_4 fibers consist of abundant tiny MgFe_2O_4 nanocrystals.

Fig. 5 shows the TEM images of $\text{Mg}_2\text{Fe}_2\text{O}_5$ fibers. The BF TEM image in Fig. 5(a) demonstrates microstructural characteristics similar to those of MgFe_2O_4 , with the fibers composed of small nanoparticles. The SAED pattern in Fig. 5(b) exhibits clear polycrystalline rings, indexed sequentially from inner to outer as the (200), (110), (011), and (020) planes of orthorhombic $\text{Mg}_2\text{Fe}_2\text{O}_5$. The HRTEM image of a typical grain in Fig. 5(c) indicates a grain size of approximately 20 nm. In Fig. 5(d), the enlarged HRTEM image of the white-boxed region in Fig. 5(c), the interplanar spacings are measured to be 3.67 Å and 3.32 Å with an angle of 90°, which can be assigned to the (011) and (400) planes of orthorhombic $\text{Mg}_2\text{Fe}_2\text{O}_5$. Thus, the $\text{Mg}_2\text{Fe}_2\text{O}_5$ submicron fibers are also composed of numerous minute $\text{Mg}_2\text{Fe}_2\text{O}_5$ nanoparticles. Collectively, these TEM observations highlight that both single-phase fibers maintain a polycrystalline nanofibrous architecture built from nanocrystals.

Extensive HRTEM examinations show that 65–70% of the interfaces are heterointerfaces in $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$ composite fibers, one of which is shown in Fig. 6. Fig. 6(a) presents a BF TEM image of the $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$ composite fibers, revealing that they are composed of abundant nanoparticles, analogous to the microstructural features of MgFe_2O_4 and $\text{Mg}_2\text{Fe}_2\text{O}_5$ fibers. The corresponding SAED pattern in Fig. 6(b) displays polycrystalline diffraction rings characteristic of both MgFe_2O_4 and $\text{Mg}_2\text{Fe}_2\text{O}_5$ phases, confirming their coexistence within the same fiber. Typical HRTEM image in Fig. 6(c) clearly reveals a high density of grain boundaries throughout the fiber. For the grain I in Fig. 6 (d), the interplanar spacing is 2.96 Å, indexed to the (220) plane of cubic

MgFe_2O_4 ; For the grain II in Fig. 6(e), two adjacent planes exhibit lattice spacings of 2.59 Å with an interplanar angle of 90°, corresponding to the (002) and (020) planes of orthorhombic $\text{Mg}_2\text{Fe}_2\text{O}_5$. In Fig. 6(f), the enlarged HRTEM image of the interface between MgFe_2O_4 and $\text{Mg}_2\text{Fe}_2\text{O}_5$ demonstrates excellent lattice matching across the boundary. This coherent or semi-coherent heterointerface likely strengthens interfacial bonding and contributes positively to the overall magnetic properties of the composite.

Fig. 7(a) provides a low-magnification overview, further illustrating that this fiber comprises of many minute crystallites. Elemental mapping in Fig. 7(b–d) shows uniform spatial distribution of three elements (Fe, O, and Mg) within the fiber. This homogeneous elemental distribution implies intimate and uniform mixing of MgFe_2O_4 and $\text{Mg}_2\text{Fe}_2\text{O}_5$ crystallites at the nanoscale, thereby maximizing the density of heterointerfaces within the composite fiber. These heterointerfaces hold promise for enhanced functional performance, particularly in magnetism.

To further elucidate the heterointerface structure within the $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$ composite fibers, STEM characterization was performed, as presented in Fig. 8. Fig. 8(a) shows an HAADF image obtained from the heterointerface region of the composite fiber. Two grains with distinct orientations are clearly visible and labeled as grain I and grain II, with the interfacial region between grain I and II denoted as region III. In Fig. 8(b), the enlarged HAADF image of grain I reveals a measured interplanar spacing of 2.59 Å, which corresponds to the (020) plane of orthorhombic $\text{Mg}_2\text{Fe}_2\text{O}_5$. In Fig. 8(c), the enlarged HAADF image of grain II displays two adjacent lattice planes with spacings of 2.96 Å and 2.53 Å, and an interplanar angle of approximately 65°, which are indexed as the (2 2̄ 0) and (1̄ 1̄ 3) planes of cubic MgFe_2O_4 , respectively. In Fig. 8(d), the enlarged HAADF image of the interface between

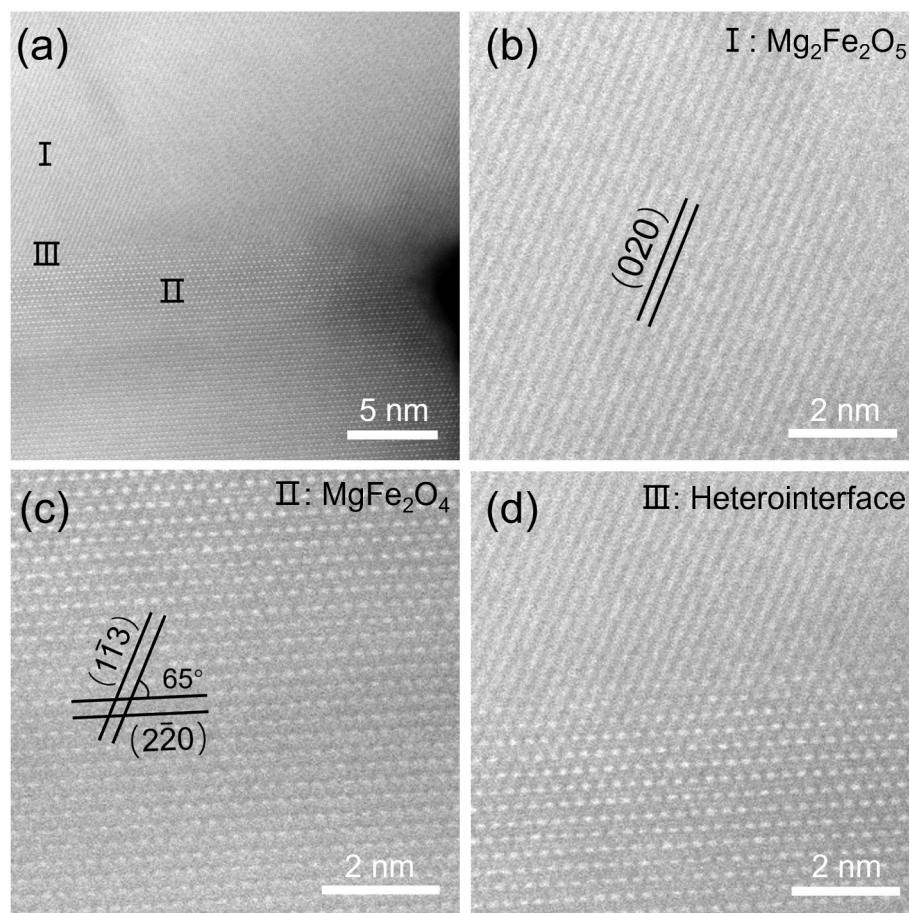


Fig. 8. HAADF images of the $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$ heterostructure interface. (a) HAADF image of the interface, (b) Enlarged HAADF image of grain I, (c) Enlarged HAADF image of grain II, (d) Enlarged HAADF image of region III.

grain I and grain II clearly reveals a sharp boundary between the MgFe_2O_4 and $\text{Mg}_2\text{Fe}_2\text{O}_5$ phases. Notably, the lattice planes across the interface exhibit an epitaxial orientation relationship: $[110] \text{MgFe}_2\text{O}_4 // [100] \text{Mg}_2\text{Fe}_2\text{O}_5$, $(1\bar{1}3) \text{MgFe}_2\text{O}_4 // (020) \text{Mg}_2\text{Fe}_2\text{O}_5$. These observations provide direct evidence for the formation of coherent or semi-coherent heterointerfaces in the $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$ composite fibers. These STEM-HAADF observations confirm the presence of well-matched heterointerfaces at the nanoscale, which are expected to play a critical role in modulating the magnetic coupling and overall functional performance of the biphasic composite fibers.

3.3. Magnetic properties of magnesium-based ferrite fibers

To investigate the magnetic properties of the magnesium-based ferrite fibers and the specific influence of the heterointerfaces on their magnetism, the hysteresis loops of MgFe_2O_4 , $\text{Mg}_2\text{Fe}_2\text{O}_5$, and $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$ fibers were measured at room temperature. The field-dependent magnetization behavior was recorded, yielding the hysteresis loops as shown in Fig. 9. From Fig. 9(a), it can be seen that all three ferrite fibers exhibit distinct soft-magnetic characteristics, evidenced by relatively low coercivity (H_c) values. The measured coercivities are 21.88 Oe for MgFe_2O_4 , 42.86 Oe for $\text{Mg}_2\text{Fe}_2\text{O}_5$, and 61.33 Oe for the $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$ composite. The presence of heterointerfaces in $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$ induces a remarkable increase in coercivity, indicating an enhanced magnetic pinning or anisotropy at the boundaries. More strikingly, the heterointerfaces exert a pronounced positive effect on magnetization. The M_s of $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$ composite fibers is significantly higher than that of single-phase MgFe_2O_4 and $\text{Mg}_2\text{Fe}_2\text{O}_5$ fibers. Close inspection of the enlarged hysteresis loops in Fig. 9(b)

further reveals that the remnant magnetization (M_r) of the composite is also superior to that of the single-phase counterparts. Detailed magnetic parameters, including H_c , M_s , and M_r , are summarized in Table 2. These findings demonstrate that construction of heterointerfaces between MgFe_2O_4 and $\text{Mg}_2\text{Fe}_2\text{O}_5$ represents an effective approach to substantially enhance the overall magnetization of magnesium ferrites, offering a promising route for tailoring soft-magnetic performance in 1D nanostructures.

The ratio of remnant magnetization to saturation magnetization (M_r/M_s) serves as a key indicator of the ability of materials to retain magnetization after removal of the external magnetic field, often referred to as magnetic squareness or remanence ratio. Among the three ferrites, the $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$ composite fibers exhibit the highest M_r/M_s value. This demonstrates that the strategic incorporation of heterointerfaces not only enhances saturation magnetization and coercivity but also significantly improves the capacity of the composite fibers to maintain a magnetized state in the absence of an applied field. It is noteworthy that both H_c and M_s are theoretically linked to the magnetic anisotropy constant (K) according to the well-established relationship given by Equation (1) [56]:

$$H_c = \frac{0.96 \times K}{M_s} \quad (1)$$

By substituting the experimental data into Equ.1, the K for the MgFe_2O_4 , $\text{Mg}_2\text{Fe}_2\text{O}_5$ and $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$ fibers were calculated to be 8.85, 88.84 and 253.63 Oe-emu/g, respectively. This indicates that the orthorhombic $\text{Mg}_2\text{Fe}_2\text{O}_5$ exhibits a higher K value than the cubic MgFe_2O_4 . This difference is likely due to the lower symmetry of the orthorhombic crystal structure. Notably, the $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$

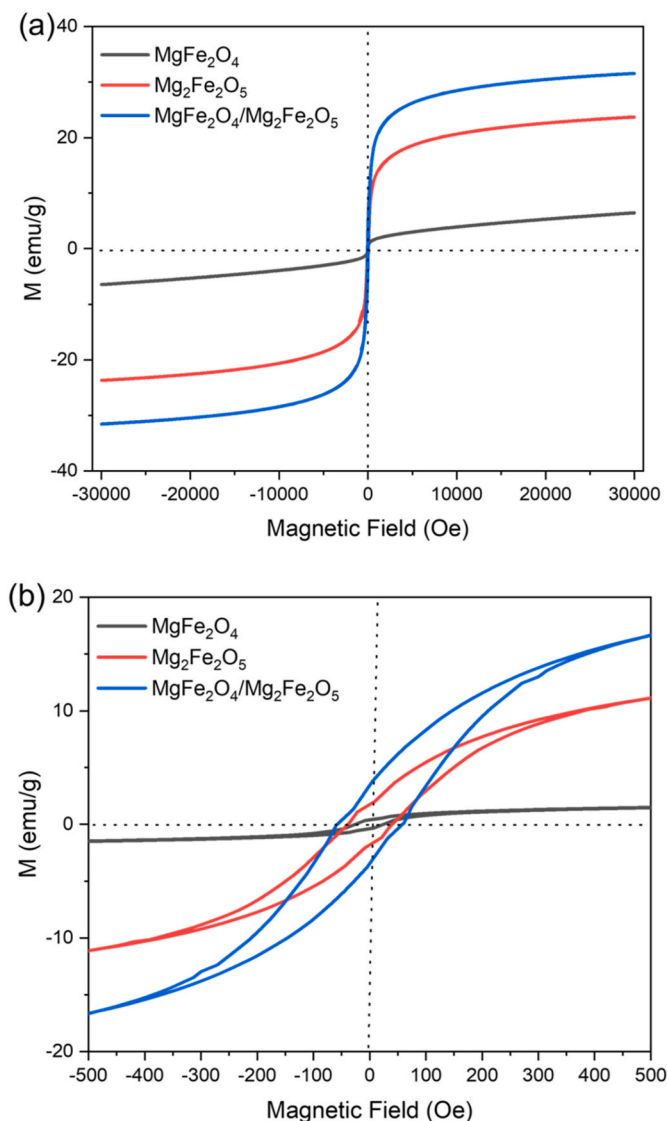


Fig. 9. (a) Magnetic hysteresis loops of MgFe_2O_4 , $\text{Mg}_2\text{Fe}_2\text{O}_5$, and $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$ fibers measured at room temperature; (b) Magnified view near the zero-field region.

Table 2
Magnetic parameters of MgFe_2O_4 , $\text{Mg}_2\text{Fe}_2\text{O}_5$, and $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$ fibers.

Materials	H_c (Oe)	M_r (emu/g)	M_s (emu/g)	M_r/M_s	K (Oe•emu/g)
MgFe_2O_4	21.88	0.39	6.46	0.06	8.85
$\text{Mg}_2\text{Fe}_2\text{O}_5$	42.86	1.99	23.72	0.08	88.84
$\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$	61.33	3.97	31.54	0.13	253.63

composite fibers yield the highest K value, significantly surpassing both single-phase fibers. This pronounced enhancement results from the abundance of heterointerfaces between the cubic and orthorhombic phases, which significantly increase the magnetic anisotropy constant of the composite fibers.

In addition to the fundamental understanding of the heterointerface effect, the substantial enhancement in saturation magnetization brings significant practical implications. For the high-frequency power inductors and electromagnetic interference filters, a higher M_s can be directly translated to higher maximum energy storage density and a

reduced component volume under the same power rating. Furthermore, an increased magnetic anisotropy constant ($K = 253.63$ Oe•emu/g) observed in the composite fibers is beneficial for suppressing magnetic relaxation losses at elevated frequencies, which is a critical requirement for 5G communication modules operating in the gigahertz range. Meanwhile, the one-dimensional fibrous morphology with a large aspect ratio facilitates easy alignment during device fabrication and provides excellent mechanical flexibility for flexible electronics. Therefore, the heterointerface engineering strategy demonstrated in this work not only effectively remedies the long-standing drawback of insufficient saturation magnetization in magnesium-based ferrites but also offers a scalable pathway toward fabricating high-performance, lightweight, and miniaturized magnetic components for next-generation electronic systems.

3.4. Calculations of electronic structure and magnetic moments in magnesium ferrites

To elucidate the underlying mechanism for the enhanced magnetic performance of the composite fibers, first-principles calculations were employed to systematically investigate the magnetic structures and Fe atomic magnetic moments in the single-phase MgFe_2O_4 and $\text{Mg}_2\text{Fe}_2\text{O}_5$,

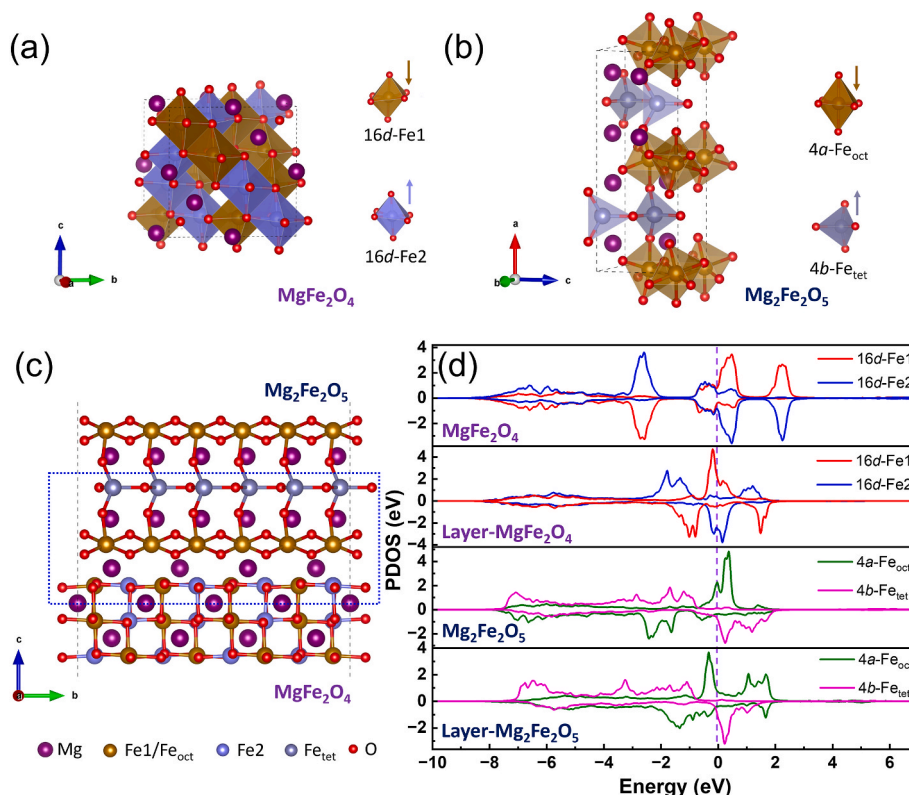


Fig. 10. (a) Unit cell of MgFe_2O_4 ; (b) Unit cell of $\text{Mg}_2\text{Fe}_2\text{O}_5$; (c) Schematic illustration of the $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$ heterointerface structure; (d) PDOS of Fe atoms in MgFe_2O_4 , $\text{Mg}_2\text{Fe}_2\text{O}_5$, and at the $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$ heterointerface.

Table 3

Magnetic moments of Fe atoms in the unit cell of MgFe_2O_4 , $\text{Mg}_2\text{Fe}_2\text{O}_5$, and at the $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$ heterointerface.

Material	Site	Atom	M(μ_B)	Average M(μ_B)
MgFe_2O_4	16d	Fe1	-3.063	0.000
	16d	Fe2	3.063	
$\text{Mg}_2\text{Fe}_2\text{O}_5$	4a	Fe_{oct}	-2.525	0.561
	4b	Fe_{tet}	3.647	
$\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$	16d	Fe1	-0.878	0.714
	16d	Fe2	1.640	
	4a	Fe_{oct}	-1.609	
	4b	Fe_{tet}	3.701	

as well as in the $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$ heterointerface model. Spin-polarized calculations were first performed on the single-phase MgFe_2O_4 and $\text{Mg}_2\text{Fe}_2\text{O}_5$ systems to identify their lowest-energy stable spin configurations, as illustrated in Fig. 10(a) and (b), respectively. Subsequently, a $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$ heterointerface model was constructed based on these stable spin structures, and then structural optimization was carried out to determine the equilibrium interfacial configuration as shown in Fig. 10(c). On this basis, the PDOS and magnetic moments of Fe atoms in all three systems were further computed, as shown in Fig. 10(d). In the MgFe_2O_4 unit cell, as shown in Fig. 10(a), Fe atoms occupy octahedral sites with two distinct spin orientations, labeled 16d-Fe1 and 16d-Fe2, exhibiting antiparallel magnetic moments, $-3.063 \mu_B$ for 16d-Fe1 and $+3.063 \mu_B$ for 16d-Fe2, resulting in a net average magnetic moment of $0.000 \mu_B$ per formula unit. From the spin-polarized PDOS for MgFe_2O_4 in Fig. 10(d), it can be seen that the spin density of these two types Fe atoms demonstrates a perfectly antiparallel distribution, consistent with the calculated magnetic moments, typical of antiferromagnetic behavior.

In $\text{Mg}_2\text{Fe}_2\text{O}_5$ unit cell shown in Fig. 10(b), Fe atoms occupy both octahedral (Fe_{oct} , labeled 4a- Fe_{oct}) and tetrahedral (Fe_{tet} , labeled 4b-

Fe_{tet}) sites, again with antiparallel alignment. The computed magnetic moments are $-2.525 \mu_B$ for 4a- Fe_{oct} and $+3.647 \mu_B$ for 4b- Fe_{tet} , leading to a net average magnetic moment of $0.561 \mu_B$ (see Table 3). The net magnetic moment of 4a- Fe_{oct} and 4b- Fe_{tet} can be directly reflected by the asymmetry of spin density; the lower the symmetry, the greater the atomic magnetic moment and vice versa. From the PDOS of $\text{Mg}_2\text{Fe}_2\text{O}_5$, the occupied PDOS of spin-up is lower than that of spin-down in 4a- Fe_{oct} , while spin-up states are higher than spin-down in 4b- Fe_{tet} . Thus, there is a relatively larger spin-up net magnetic moment in 4b- Fe_{tet} , leading to a positive net magnetic moment in the $\text{Mg}_2\text{Fe}_2\text{O}_5$ unit cell. Compared with the single-phase systems, the PDOS in Fig. 10(d) reveals a significant change of electronic states at the $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$ heterointerface. In the MgFe_2O_4 layer, two types of Fe atoms are located in the same layer at the interface, and their PDOS move towards the Fermi level. Spin-down 16d-Fe1 and spin-up 16d-Fe2 undergo spin reversal near the Fermi level, with 16d-Fe2 reversal less pronounced than 16d-Fe1, resulting in a positive net magnetic moment in the MgFe_2O_4 layer. In the $\text{Mg}_2\text{Fe}_2\text{O}_5$ layer, the spin-down states of 4a- Fe_{oct} at the first interface layer decrease substantially and the spin-up states of 4b- Fe_{tet} near the interface remain nearly unchanged, compared with those in $\text{Mg}_2\text{Fe}_2\text{O}_5$, leading to a positive net magnetic moment in the $\text{Mg}_2\text{Fe}_2\text{O}_5$ layer. Consequently, the average net magnetic moment of the heterointerface system rises significantly to $0.714 \mu_B$, exceeding those of two single-phase systems. Therefore, the $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$ heterointerfaces exert a more pronounced enhancement on the magnetic properties of Mg-based ferrite materials. Such enhancement of magnetic properties of Fe atoms at the heterointerface can also be found in FeSi@BN core-shell structure [57].

4. Conclusion

In summary, MgFe_2O_4 , $\text{Mg}_2\text{Fe}_2\text{O}_5$ and $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$ composite fibers have been prepared using electrospinning followed by high-temperature annealing. HRTEM and STEM observations show that all

three types of fibers are composed of nanocrystals, with the composite fibers containing plenty of heterointerfaces. Hysteresis loop measurements demonstrate that the saturation magnetization of the $\text{MgFe}_2\text{O}_4/\text{Mg}_2\text{Fe}_2\text{O}_5$ composite fibers is significantly higher than that of single-phase MgFe_2O_4 and $\text{Mg}_2\text{Fe}_2\text{O}_5$, indicating that the presence of heterointerfaces can effectively improve the magnetic performance of magnetite ferrites. The underlying mechanism is further elucidated by first-principles calculations, which reveal that the interface-induced reduction in spin-down electron density of interfacial Fe atoms is responsible for the increased net magnetic moment. This study not only elucidates the microscopic mechanism of interfacial magnetic coupling but also demonstrates that heterointerface engineering is an effective and universal strategy for overcoming the intrinsic limitations of soft magnetic ferrites, which can be broadly applied to other metal oxide systems.

CRediT authorship contribution statement

Zhibin Ling: Writing – original draft, Investigation, Formal analysis, Data curation, Conceptualization. **Feiyu Diao:** Writing – review & editing, Supervision, Resources. **Fei Xie:** Resources, Investigation. **Rongsheng Cai:** Validation, Data curation. **Xuyan Xue:** Supervision, Software. **Yiqian Wang:** Writing – review & editing, Supervision, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.matchemphys.2026.133144>.

Data availability

Data will be made available on request.

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