Electrospun Flexible Coaxial Nanoribbons Endowed With Tuned and Simultaneous Fluorescent Color-Electricity-Magnetism Trifunctionality

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In order to develop new-typed multifunctional nanocomposites, fluorescent-electrical-magnetic trifunctional coaxial nanoribbons with tunable fluorescent color, including white-light emission, have been successfully fabricated via coaxial electrospinning technology. Each stripe of coaxial nanoribbon is composed of a Fe$_3$O$_4$/PMMA core and a [Eu(BA)$_3$phen+Dy(BA)$_3$phen]/PANI/PMMA (PMMA = polymethyl methacrylate, BA = benzoic acid, phen = phenanthroline, PANI = polyaniline) shell. X-ray diffractometry (XRD), field emission scanning electron microscopy (FE-SEM), biological microscopy (BM), vibrating sample magnetometry (VSM), energy dispersive spectrometry (EDS), Hall effect measurement system and photoluminescence (PL) spectroscopy were employed to characterize the coaxial nanoribbons. Emitting color of the coaxial nanoribbons can be tuned by adjusting the contents of Dy(BA)$_3$phen, Eu(BA)$_3$phen, PANI and Fe$_3$O$_4$ in a wide color range of blue-white-orange under the excitation of 273-nm single-wavelength ultraviolet light. The coaxial nanoribbons simultaneously possess excellent luminescent performance, electrical conduction and magnetism compared with the counterpart composite nanoribbons. Furthermore, the electrical and magnetic performances of the coaxial nanoribbons also can be tunable by adding different quantities of PANI and Fe$_3$O$_4$ nanoparticles, respectively. The obtained coaxial nanoribbons have promising applications in many areas, such as electromagnetic interference shielding, microwave absorption, molecular electronics, biomedicine, future nanomechanics and display fields.

One dimensional (1D) multifunctional nanocomposites that possess desirable properties in a single entity have attracted broad interest in recent years. They have bi- or trifunction such as electrical, optical, magnetic and chemical properties, leading to the wide range of technological applications in various fields. Among these nanocomposites, the magnetic-fluorescent bifunctional nanocomposites combined magnetic with fluorescent functionalities have particularly attracted great attention due to the unique properties associated with promising applications, such as targeted drug delivery, magnetic resonance imaging (MRI), optical imaging, etc. Electromagnetically functionalized micro/nanostructures of conducting polymers are of special interest due to their potential applications in areas such as electromagnetic interference shielding, microwave absorption, nonlinear optics and biomedicine. Previously, polyaniline (PANI) nanotubes containing Fe$_3$O$_4$ nanoparticles (NPs) have been fabricated by an in situ chemical oxidation polymerization in the presence of Fe$_3$O$_4$ NPs. Recently, Zhang, et al. developed a very simple
chemical one-step method to prepare PANI/α-Fe₂O₃ nanofibers. Luminescent-electrical materials are expected to possess many potential applications in areas such as color display, electromagnetic shielding, molecular electronics and biomedicine. M. Ghoswami, et al. synthesized polyaniline-cadmium sulfide nanocomposite using chemical oxidative method. This composite can be used in different optoelectronic purposes and it is a promising material with prospect of application in polymer light emitting diodes (PLED). Lun, et al. fabricated novel [Tb(BA)_3phen+Eu(BA)_3phen]/PANI/PVP composite nanofibers with tunable color-electricity bifunctionality via facile one-pot electrospinning technology. The prepared composite nanofibers have potential applications in many fields such as color display, molecular electronics and biomedicine.

It is therefore of considerable interest to develop the luminescence-electrical-magnetic tri-functional nanomaterials which have potential applications in molecular electronics, biomedicine, microwave absorption, electromagnetic shielding, etc. At present, some preparations of tri-functional photoluminescence-electricity-magnetism 1D nanomaterials have been reported. Yu, et al. illustrated a route for fabricating Fe₃O₄@Au/PANI multifunctional nanocomposites with a core-shell structure, where the Fe₃O₄@Au nanoparticles are well dispersed in the PANI matrix. The obtained Fe₃O₄@Au/PANI nanocomposites exhibit integrated optical, electrical and magnetic properties. Lun, et al. prepared the tuned electricity, magnetism and fluorescent color tri-functional composite microbelts via one-pot electrospinning technique. The composite microbelts suffered heavy losses in fluorescent intensity, because Fe₃O₄ NPs were directly mixed with the RE luminescent compounds. From that study, the multifunctional composite nanomaterials suffered heavy losses in fluorescent intensity when Fe₃O₄ NPs were directly blended with the luminescent compounds. So rare earth complex must be effectively isolated from Fe₃O₄ NPs to ultimately reduce the impact of Fe₃O₄ NPs on the fluorescent property if the strong luminescence is achieved.

Among various luminescent materials, rare earth complexes doped inorganic/organic hybrids have good thermal and mechanical stabilities and processing property. Furthermore, they have excellent luminescent property on account of the antenna effect of ligands and the f-f electron transition of RE ions. Nowadays, tunable multicolor, especially white-light emissions, can be successfully achieved via varying the doping ions and doping concentration. For example, Sun, et al. prepared Gd₃Ga₅O₁₂: Yb³⁺/Tm³⁺/Ho³⁺ nanocrystals via a citric acid complex procedure. By means of adjusting the doping concentrations of Yb³⁺/Tm³⁺/Ho³⁺, the red-green-blue up-conversion luminescence was obviously changed. Yi, et al. synthesized NaCeF₄: Ln³⁺ (Eu³⁺, Dy³⁺, Tb³⁺) nanorods with tunable multicolor output and bright white emissions via a typical hydrothermal method using oleic acid as the capping agent. Guan, et al. successfully synthesized multicolor emitting Tb³⁺ and/or Sm³⁺ doped NaGdF₄ luminescent nanomaterials via an SDS-assisted one-step hydrothermal method. The as-prepared Tb³⁺ or Sm³⁺ doped samples showed strong green and yellow emission. Lun, et al. reported tunable fluorescent color-electricity bifunctional composite nanofibers prepared by electrospinning, and the emitting color of the composite nanofibers can be tuned in a wide color range of red-yellow-green by adjusting the mass ratio of terbium complexes and europium complexes, or by changing PANI content. More importantly, Guan, et al. fabricated a series of tunable luminescence NaGdF₄: Dy³⁺, Eu³⁺ nanophosphors for the first time by a one-step hydrothermal method, and the products achieved tunable multicolor and warm white-light emission under the excitation of the 273-nm ultraviolet light.

Nanoribbon is a kind of nanomaterial of special morphology. An increasing number of scientists have shown strong interest to research it owing to its anisotropy, large width-thickness ratio, unique optical, electrical and magnetic properties.

In this paper, we report assembling tunable fluorescent color-electricity-magnetism trifunctionality into [Fe₃O₄/PMMMA]@[Dy(BA)_3phen+Eu(BA)_3phen]/PANI/PMMMA] coaxial nanoribbons. Of the coaxial nanoribbons, the core is composed of template PMMA containing Fe₃O₄ NPs, and the shell consists of PMMA containing RE (Dy, Eu) complexes, or by changing PANI content. The multifunctional one-dimensional nanoribbons. The structure, fluorescence, electricity and magnetism of the coaxial nanoribbons were systematically studied, and some new results were obtained.

**Materials and Methods**

**Chemicals.** Methylmethacrylate (MMA), benzoylperoxide (BPO), Eu₂O₃ (99.99%), Dy₂O₃ (99.99%), benzoic acid (BA), 1,10-phenanthroline (phen), FeCl₃·6H₂O, FeSO₄·7H₂O, NH₄NO₃, polyethylene glycol (PEG, Mr≈20 000), ammonia, anhydrous ethanol, CHCl₃ and dimethylformamide (DMF) were bought from Tianjin Tiantai Fine Chemical Co., Ltd. Anhydrous ethanol, aniline (ANI), (IS)-(+) -Camphor-10-sulfonic acid (CSA) and oleic acid (OA) were purchased from Sinopharm Chemical Reagent Co., Ltd. Ammonium persulfate (APS) was bought from Guangdong Xilong Chemical Co., Ltd. Nitric acid (HNO₃, AR) was purchased from Beijing Chemical Works. All the reagents were of analytical grade and directly used as received without further purification. Deionized water was made in our own laboratory.
Synthesis of Eu(BA)₃phen and Dy(BA)₃phen complexes. Eu(BA)₃phen powders were synthesized according to the traditional method as described in the reference. 1.7600 g of Eu₂O₃ was dissolved in an amount of concentrated nitric acid and then crystallized via evaporation of excess nitric acid and water by heating, and Eu(NO₃)₃·6H₂O was acquired. Eu(NO₃)₃ ethanol solution was prepared by adding amount of anhydrous ethanol into the above Eu(NO₃)₃·6H₂O. 3.6640 g of BA and 1.8020 g of phen were dissolved in ethanol. The Eu(NO₃)₃ ethanol solution was then added dropwise into the mixture solution of BA and phen with magnetic agitation at 60 °C for 3 h. The precipitates were collected by filtration and washed for three times using ethanol, and then dried in an electric oven at 60 °C for 12 h. The synthetic method of Dy(BA)₃phen complex was similar to the above method, except that the using dosages of Dy₂O₃, BA and phen were 1.8650 g, 3.6600 g and 1.8000 g, respectively.

Preparation of PMMA. PMMA used in this study was prepared by oxidative polymerization of MMA (MMA (100 mL) and BPO (0.1000 g) were mixed in a 250 mL three-necked flask with a backflow device and stirred vigorously at 90–95 °C. When the viscosity of the solution reached a certain value just like that of glycerol, the heating was stopped and it was left to naturally cool down to room temperature. The obtained gelatinous solution was then loaded into test tubes, and the influx height was 5–7 cm. After that, the tubes were put in an electric vacuum oven for 48 h at 60 °C, and the gelatinous solution was then solidified. Finally, the temperature in the oven was raised to 110 °C for 2 h to terminate the reaction. The weight-average molecular weight and the degree of polymerization (DP) value of as-prepared PMMA were 9.7 × 10⁴ and 9.7 × 10⁴, respectively.

Preparation of oleic acid modified Fe₃O₄ NPs. Fe₃O₄ NPs were obtained via a facile coprecipitation synthetic method, and PEG was used as the protective agent to prevent the particles from aggregating. One typical synthetic procedure was as follows: 5.4060 g of FeCl₃·6H₂O, 2.7800 g of FeSO₄·7H₂O, 4.0400 g of NH₄NO₃ and 1.9000 g of PEG were added into 100 mL of deionized water to form a uniform solution under vigorous stirring at 50 °C. To prevent the oxidation of Fe²⁺, the reactive mixture was kept under argon atmosphere. After the mixture had been bubbled with argon for 30 min, 0.1 mol·L⁻¹ of NH₃·H₂O was dropwise added into the mixture until the pH value was above 11. Then the system was continuously bubbled with argon for 20 min at 50 °C, and black precipitates were formed. The precipitates were collected from the solution by magnetic separation, washed for three times with deionized water, and then dried in an electric vacuum oven at 60 °C for 12 h. The as-prepared Fe₃O₄ NPs were then coated with oleic acid (OA) as below: 2.0000 g of the as-prepared Fe₃O₄ NPs were ultrasonically dispersed in 100 mL of deionized water for 20 min. The suspension was heated to 80 °C under argon atmosphere with vigorous mechanical stirring for 30 min and then 1 mL of OA was dropwise added. Reaction was stopped after heating and stirring the mixture for 40 min. The precipitates were collected from the solution by magnetic separation, washed with ethyl alcohol for three times, and then dried in an electric vacuum oven for 6 h at 60 °C.

Preparations of spinning solutions for fabricating coaxial nanoribbons. Two different kinds of spinning solutions were prepared to fabricate coaxial nanoribbons. The spinning solution for the shell (denoted as spinning solution I) of coaxial nanoribbons was composed of Dy(BA)₃phen, Eu(BA)₃phen, PANI, PMMA, DMF and CHCl₃, and detailed preparation process for spinning solution I was as following: certain amount of ANI and CSA, and 0.5 g of PMMA were dissolved in the mixed solution of 0.3 g of DMF and 6 g of CHCl₃ with magnetic stirring for 48 h at room temperature (defined as solution A). Meanwhile, APS was used as an oxidant and dispersed into a mixed solution of 0.6 g of DMF and 3 g of CHCl₃ with magnetic stirring for at least 2 h at room temperature (defined as solution B). Then solution A and B were both cooled down to 0 °C in an ice-bath. Subsequently, solution B was added dropwise into solution A under magnetic stirring. The final mixture was allowed to react at 0 °C for 24 h to produce PANI by the polymerization of aniline. Then certain amounts of Eu(BA)₃phen and Dy(BA)₃phen complexes were added into the mixture under magnetic stirring for another 12 h at room temperature, thus spinning solution I for the shell was prepared.

Compared to Eu(BA)₃phen, Dy(BA)₃phen has weaker luminescence intensity. Therefore, in this study, we need firstly find the optimum concentration of Dy(BA)₃phen in the shell to guarantee the luminescence intensity of Dy(BA)₃phen to reach maximum, and then different amounts of Eu(BA)₃phen are introduced into the shell to realize tunable color. In order to find the optimum concentration of Dy(BA)₃phen, a series of Dy(BA)₃phen/PANI/PMMA composite nanoribbons were fabricated. For performing this study, the mass percentage of ANI to PMMA was settled as 30%, the mass percentages of Dy(BA)₃phen to PMMA were varied from 120% to 240%. The corresponding samples were marked as a, b, c, d and e. The compositions and contents of these composite nanoribbons were listed in Table 1. Based on photoluminescence analysis discussed in section “Photoluminescence property”, the mass percentage of Dy(BA)₃phen to PMMA settled as 180% was adopted to prepare [Fe₃O₄/PMMA]@[{Dy(BA)₃phen+Eu(BA)₃phen}/PANI/PMMA] coaxial nanoribbons. The dosages of materials used for preparing spinning solution I were shown in Table 2.

The other spinning solution for the core of coaxial nanoribbons consisted of certain amounts of OA modified Fe₃O₄ NPs, 0.5 g of PMMA, 9 g of CHCl₃ and 0.9 g of DMF (denoted as spinning solution II). Fe₃O₄ NPs were dispersed in DMF and CHCl₃ with the assist of ultrasonics for 15 min, and then
PMMA was added into the above solution under mechanical stirring. In order to investigate the impact of Fe$_3$O$_4$ NPs on the properties of coaxial nanoribbons, various contents of Fe$_3$O$_4$ NPs were introduced into spinning solution II. The compositions and contents of the spinning solutions were summarized in Table 3. The obtained coaxial nanoribbons were denoted as S$_{ax}$@S$_{by}$ (x = 1–9; y = 1–3) according to the corresponding spinning solution I and II.

### Preparations of tunable multicolor and white-light emissions luminescent-electrical-magnetic trifunctional coaxial nanoribbons.

A homemade coaxial electrospinneret was used in this study. The equipment for the electrospinning process is presented in Fig. 1. Spinning solution I was loaded into the outer plastic syringe while spinning solution II was loaded into the inner plastic syringe. A piece of flat iron net was used as a collector and put about 10 cm away from the nozzle tip. The positive terminal of a direct current (DC) high voltage power supply was connected to the carbon electrode which was immersed into the spinning solution II, and the negative terminal was connected to the iron net. Positive DC voltage of 6 kV was applied between the nozzle and the collector to generate coaxial nanoribbons under the ambient temperature of 20–25 °C, and the relative humidity of 45–50%.

### Fabrication of Fe$_3$O$_4$/[Dy(BA)$_3$phen$+$Eu(BA)$_3$phen]/PANI/PMMA composite nanoribbons.

In order to highlight advantages of coaxial nanostructure, Fe$_3$O$_4$/[Dy(BA)$_3$phen$+$Eu(BA)$_3$phen]/PANI/PMMA composite nanoribbons were also fabricated by mixing the spinning solution I-S$_{a3}$ and spinning solution II-S$_{b1}$ together at the volume ratio of 1:1. Preparation method is as follows: PANI was also obtained by the polymerization of 0.15 g of ANI, then 0.0025 g of Eu(BA)$_3$phen, 0.9 g of Dy(BA)$_3$phen

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### Table 1. Compositions of Dy(BA)$_3$phen/PANI/PMMA composite nanoribbons.

| Samples | Dy(BA)$_3$phen/g | PMMA/g |
|---------|----------------|--------|
| a       | 0.60           | 0.5    |
| b       | 0.75           | 0.5    |
| c       | 0.90           | 0.5    |
| d       | 1.05           | 0.5    |
| e       | 1.20           | 0.5    |

### Table 2. Compositions of the spinning solution I.

| Spinning solutions | Composition |
|--------------------|-------------|
| Eu(BA)$_3$phen/g   | Dy(BA)$_3$phen/g | ANI/g | CSA/g | APS/g |
| S$_{a1}$           | 0            | 0.9   | 0.15  | 0.2809 | 0.3676 |
| S$_{a2}$           | 0.0015       | 0.9   | 0.15  | 0.2809 | 0.3676 |
| S$_{a3}$           | 0.0025       | 0.9   | 0.15  | 0.2809 | 0.3676 |
| S$_{a4}$           | 0.0035       | 0.9   | 0.15  | 0.2809 | 0.3676 |
| S$_{a5}$           | 0.0150       | 0.9   | 0.15  | 0.2809 | 0.3676 |
| S$_{a6}$           | 0.0250       | 0.9   | 0.15  | 0.2809 | 0.3676 |
| S$_{a7}$           | 0.0350       | 0.9   | 0.15  | 0.2809 | 0.3676 |
| S$_{a8}$           | 0.0025       | 0.9   | 0.25  | 0.4682 | 0.6126 |
| S$_{a9}$           | 0.0025       | 0.9   | 0.35  | 0.6554 | 0.8578 |

### Table 3. Compositions of the spinning solution II.

| Spinning solutions | Composition |
|--------------------|-------------|
| Fe$_3$O$_4$/g     | PMMA/g | CHCl$_3$/g | DMF/g |
| S$_{b1}$           | 0.5    | 0.5   | 9     | 0.9   |
| S$_{b2}$           | 1.5    | 0.5   | 9     | 0.9   |
| S$_{b3}$           | 2.5    | 0.5   | 9     | 0.9   |
and 0.5 g of Fe₃O₄ NPs were added into the as-prepared PANI solution under magnetic stirring for 12 h at room temperature. The spinning voltage, distance between the collector and the spinneret and other processing parameters were the same as they were in the fabrication of the [Fe₃O₄/PMMA]@[Dy(BA)₃phen + Eu(BA)₃phen]/PANI/PMMA coaxial nanoribbons. A traditional monoaxial electrosprinneret was used to prepare the composite nanoribbons.

Characterization. The as-prepared Fe₃O₄ NPs and [Fe₃O₄/PMMA]@[Dy(BA)₃phen + Eu(BA)₃phen]/PANI/PMMA coaxial nanoribbons were examined by X-ray powder diffractometer (XRD) performed on a Bruker, D8 FOCUS diffractometer with Cu Kα radiation (λ = 0.15406 nm) and Ni filter, the operation current and voltage were maintained at 20 mA and 40 kV, and scanning speed, step length and diffraction range were settled as 10° min⁻¹, 0.1° and 20–70°, respectively. The morphology and size of Fe₃O₄ NPs were observed by a transmission electron microscope (TEM, JEOL, JEM-2010, Tokyo, Japan). The morphology and size of the coaxial nanoribbons were observed by a field emission scanning electron microscope (FESEM, XL-30) equipped with an energy-dispersive X-ray spectrometer (EDS). The internal structure of the coaxial nanoribbons was observed by a biological microscope (CVM500E). The measurements of photoluminescence (PL) spectra and the luminescence decay curves were performed by a HITACHI F-7000 fluorescence spectrophotometer using a 150 W Xe lamp as the excitation source, and scanning speed was fixed at 1200 nm·min⁻¹. The excitation and emission slits were both set to 5.0 nm. Then, the magnetic performance of Fe₃O₄ NPs, coaxial nanoribbons and composite nanoribbons was measured by a vibrating sample magnetometer (VSM, MPMS SQUID XL). The conductive property was detected by Hall effect measurement system (ECOPIA HMS-3000). The ultraviolet-visible spectra of samples were determined by a UV-1240 ultraviolet-visible spectrophotometer. All the measures were performed at room temperature.

Results and Discussion
Crystallization behavior. XRD patterns of the as-obtained Fe₃O₄ NPs, [Fe₃O₄/PMMA]@[Dy(BA)₃phen + Eu(BA)₃phen]/PANI/PMMA coaxial nanoribbons (S₃@S₇) and composite nanoribbons are shown in Fig. 2. The diffraction peaks of the as-prepared Fe₃O₄ NPs can be well indexed to the cubic phase of Fe₃O₄ (PDF 88–0315). Obviously, only characteristic peaks of Fe₃O₄ existed in the sample, and no peak from the impurities such as Fe₂O₃ and FeO(OH) could be detected, indicating the formation of pure Fe₃O₄ phase. XRD analysis results of the coaxial nanoribbons and composite nanoribbons were determined by a UV-1240 ultraviolet-visible spectrophotometer. All the measures were performed at room temperature.

Morphology and structure. The morphology of the as-prepared Fe₃O₄ NPs was observed by means of TEM, as presented in Fig. 3A. The size distribution of the spherical Fe₃O₄ NPs is almost uniform, and the particle size of the Fe₃O₄ NPs is 8.79 ± 0.085 nm (Fig. 3B). The morphology and structure of [Fe₃O₄/PMMA]@[Dy(BA)₃phen + Eu(BA)₃phen]/PANI/PMMA coaxial nanoribbons (S₃@S₇) were characterized by a combination of SEM, BM and EDS line scan analysis. SEM image of the nanoribbons is shown...
in Fig. 3C. It is found that the as-prepared nanoribbons are relatively uniform, the width of the coaxial nanoribbons is $22.52 \pm 0.153\mu m$ (Fig. 3D) and the thickness is about $2.65\mu m$.

To further confirm the coaxial structure of the \([\text{Fe}_3\text{O}_4/\text{PMMA}]@([\text{Dy}(\text{BA})_3\text{phen}]+\text{Eu}(\text{BA})_3\text{phen}]/\text{PANI}/\text{PMMA}\) coaxial nanoribbons, EDS line-scan analysis was performed, in which Dy, Eu, S and Fe elements represent Dy(BA)3phen, Eu(BA)3phen, CSA doped PANI and Fe 3O4, respectively, as shown in Fig. 3E. Elemental Fe only existed in the middle domain of the \([\text{Fe}_3\text{O}_4/\text{PMMA}]@([\text{Dy}(\text{BA})_3\text{phen}]+\text{Eu}(\text{BA})_3\text{phen}]/\text{PANI}/\text{PMMA}\) coaxial nanoribbons. The amounts of elemental Dy, Eu and S in the middle domain of the nanoribbons are lower than that in both sides of the nanoribbons because Dy(BA)3phen, Eu(BA)3phen, CSA doped PANI only exist in the top and bottom surfaces of the middle domain of the nanoribbons. It is further found that only elemental S without elemental Fe is dispersed in both sides of the coaxial nanoribbons. These results are consistent with the core-shell structure of coaxial nanoribbons.

Depending on the transmission light of the BM, the inner structure of the coaxial nanoribbons can also be observed. As revealed in Fig. 3F, a clear coaxial structure can be seen in the \([\text{Fe}_3\text{O}_4/\text{PMMA}]@([\text{Dy}(\text{BA})_3\text{phen}]+\text{Eu}(\text{BA})_3\text{phen}]/\text{PANI}/\text{PMMA}\) coaxial nanoribbons. The core of the coaxial nanoribbon is about 7\mu m in width and it contains large quantities of dark-colored \text{Fe}_3\text{O}_4\text{ NPs}, and the shell of the coaxial nanoribbon is blackish green due to the existence of PANI.

From the SEM and BM observations and EDS line-scan analysis, we can safely draw a conclusion that \([\text{Fe}_3\text{O}_4/\text{PMMA}]@([\text{Dy}(\text{BA})_3\text{phen}]+\text{Eu}(\text{BA})_3\text{phen}]/\text{PANI}/\text{PMMA}\) coaxial nanoribbons have been successfully prepared.

**Photoluminescence property.** In order to find appropriate content of Dy(BA)phen, a series of Dy(BA)phen/PANI/PMMA composite nanoribbons were fabricated by electrospinning using different spinning solutions indicated in Table 1. The excitation and emission spectra of Dy(BA)phen/PANI/PMMA composite nanoribbons are provided in Fig. 4. From the excitation spectra (Fig. 4A, left), a broad excitation band extending from 200 to 350 nm is observed from each sample when monitoring wavelength is 574 nm. The strongest peak at 273 nm assigned to the $\pi\rightarrow\pi^*$ electron transition of the ligands could also be identified. Characteristic emission peaks of the Dy(BA)phen are observed under the excitation of 273-nm ultraviolet light, which are ascribed to the energy levels transitions of $^4F_{9/2}\rightarrow^6H_{15/2}$ (481 nm) and $^4F_{9/2}\rightarrow^6H_{13/2}$ (574 nm). One can see that the photoluminescence intensity of Dy(BA)phen/PANI/PMMA composite nanoribbons is increased with adding more Dy(BA)phen. In order to further discuss the variation trend, the intensities of predominant emission peaks at 481 nm and 574 nm versus different mass percentages of Dy(BA)phen to PMMA are plotted in Fig. 4B. Obviously, the fluorescence intensity only slightly increases with introducing more Dy(BA)phen than the mass percentage of 180%. Therefore, the mass percentage of Dy(BA)phen to PMMA settled as 180% was adopted to prepare \([\text{Fe}_3\text{O}_4/\text{PMMA}]@([\text{Dy}(\text{BA})_3\text{phen}]+\text{Eu}(\text{BA})_3\text{phen}]/\text{PANI}/\text{PMMA}\) coaxial nanoribbons.

To study the color-tunable property of coaxial nanoribbons, the mass percentages of Eu(BA)phen to PMMA were varied from 0 to 5% (samples Sx@Sb1, x = 1–7), while the mass percentage of PANI to PMMA was settled as 30% and the mass ratio of Fe3O4 to PMMA was fixed as 1:1. Figure 5A shows excitation spectra of coaxial nanoribbons monitored at 574 nm, where 574 nm is the characteristic emission wavelength of Dy3+. Figure 5B demonstrates the excitation spectra of the samples monitored at 616 nm, where 616 nm is the characteristic emission wavelength of Eu3+. The strongest peak at 273 nm assigned to the $\pi\rightarrow\pi^*$ electron transition of the ligands could be also identified, and the excitation intensity is increased with introducing more Eu(BA)phen. The blue shift of excitation peak of Dy3+ is probably due to the strong absorption of Eu(BA)phen around 273 nm, resulting in the decrease of light absorption of Eu(BA)phen around 273 nm.

Figure 6A displays the emission spectra of sample Sx@Sb1 (x = 1–7). Upon excitation with 273-nm ultraviolet light, coaxial nanoribbons exhibit several main emission bands, whose positions locate at
Figure 3. TEM image (A) and histogram of particle size of Fe$_3$O$_4$ NPs (B), SEM image (C), histogram of width (D), EDS line scan analysis (E) and BM image (F) of coaxial nanoribbons (S$_{a3}$@S$_{b0}$).
Figure 4. Excitation spectra (left) and emission spectra (right) of Dy(BA)₃phen/PANI/PMMA composite nanoribbons containing different mass percentages of Dy(BA)₃phen complex (A) and relationship between Dy(BA)₃phen contents and intensities of luminescent peaks at 481 nm and 574 nm for Dy(BA)₃phen/PANI/PMMA composite nanoribbons.

Figure 5. Excitation spectra of samples Sₓ@S₃ (x = 1–7) monitored at 574 nm (A) and 616 nm (B).

Figure 6. Emission spectra of samples Sₓ@S₃ (x = 1–7) when the mass percentage of PANI is fixed as 30% and the mass ratio of Fe₃O₄ to PMMA is fixed as 1:1 (A), dependence of the emission intensity on the various samples (B).
481 nm, 574 nm, 592 nm and 616 nm. It is found that the emissions at 481 and 574 nm are due to the energy levels transitions of \(4F_{9/2} \rightarrow 6H_{13/2} \) of Dy\(^{3+}\) and the peaks at 592 and 616 nm are corresponding to the energy levels transitions of \(5D_0 \rightarrow 7F_1\) and \(5D_0 \rightarrow 7F_2\) of Eu\(^{3+}\), respectively. It is interesting and reasonable to suggest that the emission intensity of Eu\(^{3+}\) is increased, whereas that of the Dy\(^{3+}\) is simultaneously found to decrease monotonically with the increase of Eu\(^{3+}\) ions concentration. In order to clearly depict the variation trend, the intensities of the characteristic emission peaks of each sample versus different samples were plotted in the Fig. 6B. The variation of the PL intensity of the Eu\(^{3+}\) and Dy\(^{3+}\) can be attributed to the energy distribution. Since the energy that the matrix absorbs and the content of Dy(BA)\(_3\)phen are constant, more energy is assigned to Eu\(^{3+}\) with the increase of Eu(BA)\(_3\)phen content.

| Samples     | Eu(BA)\(_3\)phen concentration (n%) | CIE coordinates (x, y) |
|-------------|--------------------------------------|------------------------|
| Sa1@Sb1     | 0                                    | (0.223, 0.274)         |
| Sa2@Sb1     | 0.3                                  | (0.285, 0.308)         |
| Sa3@Sb1     | 0.5                                  | (0.320, 0.322)         |
| Sa4@Sb1     | 0.7                                  | (0.344, 0.337)         |
| Sa5@Sb1     | 1.0                                  | (0.405, 0.357)         |
| Sa6@Sb1     | 3.0                                  | (0.445, 0.353)         |
| Sa7@Sb1     | 5.0                                  | (0.598, 0.338)         |

Table 4. Comparison among the CIE chromaticity coordinates \((x, y)\) for the coaxial nanoribbons excited by 273-nm ultraviolet light.
thus leading to stronger fluorescence peaks at 592 and 616 nm. Meanwhile, on the contrary, the energy assigned to Dy\(^{3+}\) is reduced and the fluorescence peaks at 481 and 574 nm are relevantly weakened.

Generally, color can be represented by the Commission Internationale de L’Eclairage (CIE) chromaticity coordinates. The CIE chromaticity coordinates for the samples and their corresponding photographs upon excitation at 273-nm ultraviolet light are provided in Table 4 and Fig. 7. It is found that the emitting color of coaxial nanoribbons (sample S\(_{ax}@S_{b1}\) = 1–7) could be tuned by adjusting the mass ratio of Eu(BA)\(_3\)phen complexes in a wide color range of blue-white-orange. Among all the coaxial nanoribbons, the coaxial nanoribbons with 0.7% Eu\(^{3+}\) (sample S\(_{ax}@S_{b1}\)) have coordinates of x = 0.344, y = 0.337 which are close to those of standard white light (x = 0.333, y = 0.333), indicating that the as-obtained coaxial nanoribbons can emit warm white-light color. It is gratify to see that the warm white emission can be selectively realized by the co-doping of Dy(BA)\(_3\)phen and Eu(BA)\(_3\)phen complexes into coaxial nanoribbons.

The fluorescence lifetime curves of Dy\(^{3+}\) emission at 574 nm and Eu\(^{3+}\) emission at 616 nm in samples S\(_{ax}@S_{b1}\) (x = 1–7) under the excitation of 273-nm ultraviolet light are shown in Fig. 8 (A,B). From Fig. 8(A,B), one can see that all the luminescent decay lifetimes fit the single exponential rule by the following equation as the Fig. 8 (A,B) depicts.

\[
\tau = \frac{I_t}{I_0} = I_0 \exp(-t/\tau)
\]

Where \(I_t\) and \(I_0\) are the luminescence intensities at times t and 0, respectively, t is the decay time and \(\tau\) is the lifetime. Figure 8A shows that fluorescence decay lifetime of the \(^4F_{9/2} \rightarrow ^6H_{13/2}\) transitions (\(\lambda_{em} = 574\) nm) in coaxial nanoribbons is extended with increasing in Eu\(^{3+}\) concentration, while fluorescence decay lifetime of the \(^4D_{0} \rightarrow ^2F_{2}\) transitions (\(\lambda_{em} = 616\) nm) decreases, as shown in Fig. 8B. On one hand, the relative content of Dy(BA)\(_3\)phen complex in the coaxial nanoribbons is reduced with introducing more Eu(BA)\(_3\)phen. Thus the distance among Dy\(^{3+}\) in Dy(BA)\(_3\)phen molecular clusters and/or nanoparticles in the coaxial nanoribbons is increased, resulting in that the energy transfer among Dy\(^{3+}\) to Dy\(^{3+}\) is reduced and the fluorescence lifetime of Dy\(^{3+}\) is prolonged. On the other hand, more
aggregates of Eu(BA)₃phen are formed in the polymer matrix with introducing more Eu(BA)₃phen. The exciton migration between the Eu(BA)₃phen molecules shortens the fluorescence lifetime of Eu³⁺ Ref. 32.

Meanwhile, the coaxial nanoribbons containing different amounts of PANI and Fe₃O₄ NPs were fabricated to research the effect of adding different contents of Fe₃O₄ NPs (samples Sₐₙ@Sₐₚ, Sₐₚ@Sₐₙ, Sₐₚ@Sₐₙ, as illustrated in Fig. 9) and PANI (samples Sₐₙ@Sₐₚ, Sₐₚ@Sₐₙ, Sₐₚ@Sₐₙ, as shown in Fig. 10) on the fluorescent properties of the coaxial nanoribbons. As shown in Fig. 9 (A–C), the excitation and emission intensity of coaxial nanoribbons are decreased with the increase of Fe₃O₄ NPs content. Figure 9D is the CIE chromaticity coordinate diagram of coaxial nanoribbons containing different mass percentages of PANI.

Figure 10. Excitation spectra (A,B), emission spectra (C) and CIE chromaticity coordinate diagram (D) of [Fe₃O₄/PMMMA]@[Dy(BA)₃phen+Eu(BA)₃phen]/PANI/PMMMA coaxial nanoribbons containing different mass percentages of PANI.

Figure 11. Ultraviolet-visible absorption spectra of PANI/PMMMA and Fe₃O₄ NPs.

aggregates of Eu(BA)₃phen are formed in the polymer matrix with introducing more Eu(BA)₃phen. The exciton migration between the Eu(BA)₃phen molecules shortens the fluorescence lifetime of Eu³⁺ Ref. 32.

Meanwhile, the coaxial nanoribbons containing different amounts of PANI and Fe₃O₄ NPs were fabricated to research the effect of adding different contents of Fe₃O₄ NPs (samples Sₐₙ@Sₐₚ, Sₐₚ@Sₐₙ, Sₐₚ@Sₐₙ, as illustrated in Fig. 9) and PANI (samples Sₐₙ@Sₐₚ, Sₐₚ@Sₐₙ, Sₐₚ@Sₐₙ, as shown in Fig. 10) on the fluorescent properties of the coaxial nanoribbons. As shown in Fig. 9 (A–C), the excitation and emission intensity of coaxial nanoribbons are decreased with the increase of Fe₃O₄ NPs content. Figure 9D is the CIE chromaticity coordinate diagram of coaxial nanoribbons containing different Fe₃O₄ NPs contents under the excitation of 273-nm ultraviolet light. It demonstrates that the emitting color of the coaxial nanoribbons shifts with introducing more Fe₃O₄ NPs. Similarly, when the amount of PANI is increased, the excitation and emission intensity of coaxial nanoribbons are decreased as illustrated in Fig. 10 (A–C).
The CIE chromaticity coordinates for the samples and their corresponding photographs upon excitation at 273-nm ultraviolet light are provided in the Fig. 10D. It is found that the emitting color of coaxial nanoribbons could be shifted by adjusting the mass ratio of PANI.

The above results can be explained as the light absorption of Fe$_3$O$_4$ NPs and PANI. From the absorption spectra of Fe$_3$O$_4$ NPs and PANI illustrated in Fig. 11, it is seen that PANI doped PMMA strongly absorb the light in the regions of ultraviolet light ($<400$ nm) and 400–800 nm, and Fe$_3$O$_4$ NPs can absorb visible light (400–760 nm) and much more easily absorb the ultraviolet light ($<400$ nm). Thus, the exciting light and emitting light are absorbed by the PANI and Fe$_3$O$_4$ NPs, resulting in the decrease in the intensity of excitation and emission peaks. Moreover, the light absorbance becomes stronger with more PANI and Fe$_3$O$_4$ NPs introduced into coaxial nanoribbons. On the other hand, because PANI has different absorbance to different wavelengths of light, as well as Fe$_3$O$_4$ NPs, as seen in Fig. 11, different wavelengths of light emitted from coaxial nanoribbons are unequally absorbed by PANI and Fe$_3$O$_4$, leading to the fact that the emitting colors are shifted.

The fluorescent properties of coaxial nanoribbons are further investigated by comparing with that of the Fe$_3$O$_4$/[Dy(BA)$_3$phen $+$ Eu(BA)$_3$phen]/PANI/PMMA composite nanoribbons. As manifested in Fig. 12, it is found that the emission and excitation intensity of the Fe$_3$O$_4$/[Dy(BA)$_3$phen $+$ Eu(BA)$_3$phen]/PANI/PMMA composite nanoribbons is much weaker than that of the coaxial nanoribbons, and this weak fluorescent emission intensity makes the Fe$_3$O$_4$/[Dy(BA)$_3$phen $+$ Eu(BA)$_3$phen]/PANI/PMMA composite nanoribbons impractical in fluorescent performance. This heavy loss in fluorescent emission intensity is from the strong light absorption of the dark-colored Fe$_3$O$_4$ NPs when Fe$_3$O$_4$ NPs are directly blended with Dy(BA)$_3$phen and Eu(BA)$_3$phen. As illustrated in Fig. 13, since the Fe$_3$O$_4$ NPs are distributed over the whole parts of the Fe$_3$O$_4$/[Dy(BA)$_3$phen $+$ Eu(BA)$_3$phen]/PANI/PMMA composite nanoribbons, the exciting light has to pass through many Fe$_3$O$_4$ NPs to reach the Dy(BA)$_3$phen and Eu(BA)$_3$phen complexes. In this process, a large part of the exciting light has been absorbed by the Fe$_3$O$_4$ NPs, so the exciting light is much weakened before it reaches the Dy(BA)$_3$phen and Eu(BA)$_3$phen complexes.
Figure 13. Schematic diagrams of the situation of the exciting light and emitting light in the Fe$_3$O$_4$/[Dy(BA)$_3$phen+Eu(BA)$_3$phen]/PANI/PMMA composite nanoribbons and [Fe$_3$O$_4$/PMMA]@[Dy(BA)$_3$phen+Eu(BA)$_3$phen]/PANI/PMMA coaxial nanoribbons.

| Samples     | Conductivity (S·cm$^{-1}$) | Resistivity (Ω·cm) |
|-------------|-----------------------------|-------------------|
| Sa@Sb1      | $9.42 \times 10^{-4}$      | $1.062 \times 10^2$ |
| Sa8@Sb1     | $4.26 \times 10^{-3}$      | $2.35 \times 10^2$  |
| Sa9@Sb1     | $1.47 \times 10^{-2}$      | $6.8 \times 10^1$  |
| Composite nanoribbons | $6.03 \times 10^{-3}$      | $1.658 \times 10^3$ |

Table 5. Electrical conductivity and resistivity of the samples doped with various amount of PANI.

Figure 14. Hysteresis loops of Fe$_3$O$_4$ NPs, Fe$_3$O$_4$/[Dy(BA)$_3$phen+Eu(BA)$_3$phen]/PANI/PMMA composite nanoribbons and [Fe$_3$O$_4$/PMMA]@[Dy(BA)$_3$phen+Eu(BA)$_3$phen]/PANI/PMMA coaxial nanoribbons.
renders more efficient charge transport. As PANI is consecutive to Dy(BA)3phen and Eu(BA)3phen complexes, the higher electrical conductivity of the electrospun coaxial nanoribbons. As PANI is consecu-
sion of the Fe3O4/Dy(BA)3phen emission intensity of the coaxial nanoribbons is achieved by isolating Dy(BA)3phen and Eu(BA)3phen complexes. More significantly, warm white luminescent color can be achieved. It is also found that PANI and Fe3O4 NPs affect luminescent color as well. The luminescent color, magnetism and electricity trifunctionality were successfully synthesized by electrospinning technology using a specially designed coaxial spinneret. The core of the coaxial nanoribbons is composed of Fe3O4 NPs and PMMA, and the shell consists of Dy(BA)3phen, Eu(BA)3phen, PANI and PMMA. Under the excitation of 273-nm single-wavelength ultraviolet light, the emitting color of the coaxial nanoribbons is 8.38 emu·g−1 when percentages of PANI is 30 wt%, 50 wt % and 70 wt %, respectively. Obviously, the more PANI introduced into the coaxial nano-
ribbons, the higher electrical conductivity of the coaxial nanoribbons are much higher than those of the composite nanoribbons, and magnetic property compared with composite nanoribbons, while the fluorescent intensity and electrical conductivity of the coaxial nanoribbons are much higher than the counterpart composite nanoribbons. Based on the above experimental results, we can safely conclude that the shortcomings of the existing luminescence-electricity-magnetism 1D nano-
materials described in the introduction have been greatly overcome.

| Samples | Saturation magnetization (Ms) (emu·g−1) |
|---------|------------------------------------------|
| Fe3O4 nanoparticles | 42.17 |
| Composite nanoribbons | 8.38 |
| Sa3@Sb3 (Fe3O4 : PMMA = 1:1) | 7.23 |
| Sa3@Sb3 (Fe3O4 : PMMA = 3:1) | 10.8 |
| Sa3@Sb3 (Fe3O4 : PMMA = 5:1) | 18.58 |

Table 6. Saturation magnetization of Fe3O4 NPs, Fe3O4/[Dy(BA)3phen+Eu(BA)3phen]/PANI/PMMA composite nanoribbons and [Fe3O4/PMMA]@[Dy(BA)3phen+Eu(BA)3phen]/PANI/PMMA coaxial nanoribbons.

**Electrical conductivity analysis.** The conductivity of coaxial nanoribbons can be tuned by adjusting the mass percentage of PANI to PMMA. It is found from Table 5 that the electrical conductivities of products are 9.42 × 10−4 S·cm−1, 4.26 × 10−5 S·cm−1 and 1.47 × 10−2 S·cm−1 when percentages of PANI is 30 wt%, 50 wt % and 70 wt %, respectively. Obviously, the more PANI introduced into the coaxial nano-
ribbons, the higher electrical conductivity of the electrospun coaxial nanoribbons. As PANI is consecutive to the shell of coaxial nanoribbons and probably forms the conducting network more easily, which renders more efficient charge transport.

The conductivity of coaxial nanoribbon (sample Sa3@Sb1) is nearly slightly bigger than that of Fe3O4/[Dy(BA)3phen+Eu(BA)3phen]/PANI/PMMA composite nanoribbons under the same components and contents of the two kinds of nanostructures. The reason is probably that the insulative materials such as Fe3O4 NPs and RE(BA)3phen (RE = Dy, Eu) are dispersed in the Fe3O4/[Dy(BA)3phen+Eu(BA)3phen]/PANI/PMMA composite nanoribbons, which hinders the formation of continuous conductive network.

**Magnetic property.** The typical hysteresis loops for Fe3O4 NPs, Fe3O4/[Dy(BA)3phen+Eu(BA)3phen]/PANI/PMMA composite nanoribbons and [Fe3O4/PMMA]@[Dy(BA)3phen+Eu(BA)3phen]/PANI/PMMA coaxial nanoribbons containing various mass ratios of Fe3O4 NPs are shown in Fig. 14, and the saturation magnetizations of them are listed in Table 6. It is found that the saturation magnetization of the [Fe3O4/PMMA]@[Dy(BA)3phen+Eu(BA)3phen]/PANI/PMMA coaxial nanoribbons is increased with the increase of the amount of Fe3O4 NPs in the core. Besides, the saturation magnetization of the Fe3O4/[Dy(BA)3phen+Eu(BA)3phen]/PANI/PMMA composite nanoribbons is 8.38 emu·g−1 which is close to that of the coaxial nanoribbons (7.23 emu·g−1, Sa3@Sb1). By combining the analyses of magnetism, electrical conductivity and fluoroscence, it is found that the coaxial nanoribbons have close magnetic property compared with composite nanoribbons, while the fluorescent intensity and electrical conductivity of the coaxial nanoribbons are much higher than those of the composite nanoribbons, and those further demonstrate that the coaxial nanoribbons possess better luminescent-electrical-magnetic performance than the counterpart composite nanoribbons. Based on the above experimental results, we can safely conclude that the shortcomings of the existing luminescence-electricity-magnetism 1D nano-
materials described in the introduction have been greatly overcome.

**Conclusions**
[Fe3O4/PMMA]@[Dy(BA)3phen+Eu(BA)3phen]/PANI/PMMA coaxial nanoribbons with tunable fluorescent color, magnetism and electricity trifunctionality were successfully synthesized by electrospinning technology using a specially designed coaxial spinneret. The core of the coaxial nanoribbons is composed of Fe3O4 NPs and PMMA, and the shell consists of Dy(BA)3phen, Eu(BA)3phen, PANI and PMMA. Under the excitation of 273-nm single-wavelength ultraviolet light, the emitting color of the coaxial nanoribbons can be tuned in a wide color range of blue-white-orange by adjusting the mass ratio of Dy(BA)3phen and Eu(BA)3phen complexes. More significantly, warm white luminescent color can be achieved. It is also found that PANI and Fe3O4 NPs affect luminescent color as well. The luminescent intensity, electrical conductivity and magnetic property of the coaxial nanoribbons can be tunable by adjusting the contents of RE complexes, PANI and Fe3O4 NPs, respectively. It is very exciting to see that the coaxial nanoribbons simultaneously possess excellent luminescent performance, electrical conduction...
and magnetic properties. The high-performance luminescent-electrical-magnetic trifunctional coaxial nanoribbons have potential applications in molecular electronics, microwave absorption, color display device and future nanodevice.

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Author Contributions
H.S. and X.T.D. conceived and designed the study. X.T.D. supervised the research. H.S. performed the experiments. All authors discussed the results and contributed to the data interpretation. H.S. and X.T.D. wrote the manuscript and all authors commented on the manuscript.

Additional Information
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