REVIEW ARTICLE

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Organic magnetoelectric and optomagnetic couplings: perspectives for organic spin optoelectronics

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Abstract

Over the past years, the development of organic ferromagnetic materials has been investigated worldwide for potential applications. Due to the couplings among the charge, orbit, spin, and phonon in organic ferromagnetic materials, magnetoelectric, and optomagnetic couplings have been realized and observed. In this review, progress in organic magnetoelectric and optomagnetic couplings is presented, and the mechanisms behind the phenomena are also briefly summarized. Hopefully, the understanding of magnetoelectric and optomagnetic couplings could provide guidance for the further development of organic spin optoelectronics.

Introduction

Owing to the development of electronic devices toward miniaturization and high integration, optomagnetic and magnetoelectric materials are of growing interest in the fields of sensors and memory, where they allow control of the magnetization (or polarization) by an external field^{1–8}. The charge, orbit, spin, and phonon are strongly coupled, endowing ferromagnetic materials with rich coupling properties, such as magnetoelectric coupling and optomagnetic coupling^{9–12}.

In addition to inorganic materials, organic ferromagnetic materials are new candidates for the observation of magnetoelectric and optomagnetic couplings. To date, many organic magnetoelectric and optomagnetic materials have been synthesized and obtained^{13–15}. As this field has been developing very broadly and rapidly, many of the latest achievements are presented in this review, including organic ferromagnetic materials, organic magnetoelectric materials, and organic optomagnetic complexes.

Organic ferromagnetic materials

Organic materials have many incomparable advantages, such as low cost, good biological compatibility and various physical properties. Based on these excellent characteristics, the exploration of room-temperature organic ferromagnets has attracted the interest of many researchers, and great achievements have been made. In the following, we will present the progress in the development of organic ferromagnets.

Organometal molecular magnets

Molecular magnets (MMs) containing metal ions and organic components can be used as a new generation of high-density information storage devices, quantum computing devices, or spintronic devices^{16–22}. Usually, MMs have transition metal ions. The introduction of 3d and mixed 3d-4f electrons could lead to a high ground-state spin magnitude. Thus, molecules exhibit the property of a single-molecule magnet. Increasing the ground-state spin magnitude is regarded as an effective way to obtain a high effective energy barrier U_{eff} ²³. The design and synthesis of 3d-4f heteronuclear MMs have become an important research direction due to their two advantages in enhancing the energy barrier U_{eff} : first, rare earth ions can increase the magnetic anisotropy; second, the magnetic

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exchange between d and f ions can lead to a large magnitude of the magnetic moment in the ground state²⁴. Consequently, many $3d$ - $4f$ MM-based clusters were fabricated, such as Mn-Ln clusters^{25–30}, Co-Ln clusters^{31,32}, Ni-Ln clusters^{33,34}, Fe-Ln clusters^{35,36}, and Zn-Ln clusters^{37,38}.

J. Miller and coworker played a foundational role in the development of MMs. In 1991, they reported the room-temperature molecular magnet $\text{V}(\text{TCNE})_2 \cdot 1/2(\text{CH}_2\text{Cl}_2)$. This magnet had a strong ferromagnetic property at room temperature. The ferromagnetism was generated due to antiferromagnetic coupling between V and the two $[\text{TCNE}]$ -ligands³⁹. In 1993, the Mn ion-based single-molecule magnet $[\text{Mn}_{12}\text{O}_{12}(\text{O}_2\text{CMe})_{16}(\text{H}_2\text{O})_4]$ was synthesized by Sessoli et al.⁴⁰. Based on the above two discoveries, researchers synthesized many MMs using $3d$ metal ions (Mn, Fe, Co, Ni, and V) as the center ions. For Mn ion-based magnets, $[\text{Mn}_{12}\text{O}_8\text{X}_4(\text{O}_2\text{CPh})_8(\text{HMP})_6]$ possesses a novel structure with a ground-state spin magnitude of 7⁴¹. In addition, MMs with Mn_3 and Mn_4 have also been reported, for example, $[\text{Mn}_4(\text{O}_2\text{CMe})_2(\text{Hpdm})_6]^{2+}$ containing two Mn^{II} and two Mn^{III} ions. Its ground-state spin magnitude was 9⁴². In 2004, Christou et al.⁴³ reported a magnet based on clusters of Mn_{84} ions with a ring structure: $[\text{Mn}_{84}\text{O}_{72}(\text{O}_2\text{CMe})_{78}(\text{OMe})_{24}(\text{MeOH})(\text{H}_2\text{O})_{42}(\text{OH})_6] \cdot x\text{H}_2\text{O} \cdot y\text{CHCl}_3$. However, the ground-state spin magnitude was only 6. In the same year, a Mn_{25} ion-based magnetic cluster was realized by the G. Christou group that had a ground-state spin magnitude as high as 51/2. Therefore, a high Mn ion cluster number of MMs does not mean a high ground-state spin magnitude.

In addition to Mn ion-based MMs, Gatteschi et al. reported a Fe^{III}_4 cluster with a triangular structure⁴⁴. The ground-state spin magnitude was 5. In 2004, Raptopoulou

et al. obtained a butterfly-shaped Fe^{III}_4 cluster, where the ground-state spin magnitude was only 1^{45,46}. In 2010, Preowell's group prepared Fe^{III} cluster compounds that had not only a novel topological structure but also outstanding magnetic properties. The ground-state spin magnitude of this cluster could reach as high as 13/2⁴⁷. Moreover, for the cobalt ion-based MM $\text{Co}_4(\text{HMP})_4 \cdot (\text{MeOH})_4\text{Cl}_4$, four Co^{II} ions and four μ_3 -O atoms alternately occupy eight vertices of the cubic alkane. Magnetic measurements showed that its ground-state spin magnitude was 6⁴⁸. In 2015, Gao et al. reported a linear trinuclear cobalt(II) SMM⁴⁹.

Recently, Wang et al. reported the MM $[\text{Dy}(\text{L})_2(\text{C}_2\text{H}_5\text{OH})\text{Cl}_3] \cdot \text{C}_2\text{H}_5\text{OH}$ (L = tricyclohexylphosphine oxide)⁵⁰. The structure and magnetic hysteresis loops are shown in Fig. 1. Owing to the high-spin state of the Dy^{III} ion, the strong spin-lattice coupling can tune the structure of this MM, thus affecting the dielectric permittivity. This was the first time that the magnetodielectric coupling effect was explored in MMs. The relative change in the magnitude of the dielectric permittivity of this SMM was $\sim 0.75\%$ under 80 kOe, and a larger dielectric permittivity may be obtained by further increasing the applied magnetic field. The magnetodielectric coupling effect of this MM was comparable to that of some inorganic multiferroic compounds⁵¹. Overall, room-temperature MMs provide a good platform for the development of magnetoelectric coupling.

Organic ferromagnets

In addition to organic molecular magnets⁵², pure conjugated organic ferromagnets also exhibit controllable ferromagnetism^{53,54}. In 1987, by polymerizing 1,4-bis-(2,2,6,6-tetramethyl-4-oxy-4-piperidyl-1-oxy)-butadiin (BIPO), Korshak et al.⁵⁵ prepared the first polymer ferromagnet, poly-BIPO, with $T_C = 400$ K, and a superexchange model

Fig. 2 Magnetic properties of the doped P3MT. **a** M versus H curves of ClO_4^- partially doped P3MT at 5 K (squares) and 300 K (crosses). Solid line: fitting with the expression $M(H) = M_1 + M_2 \tanh(aH)$ for $H \geq 5$ kOe. Details of the ferromagnetic hysteresis. **b** Zero-field cooling dc magnetic susceptibility of ClO_4^- partially doped P3MT measured with a SQUID magnetometer. Reproduced from Nascimento et al.⁵⁸ with permission from the American Physical Society.

was proposed to explain the origin of the ferromagnetism. In the past several decades, great progress has been made in the development of polymeric ferromagnets. In 2001, Rajca et al.⁵⁶ synthesized one conjugated polymer with a large magnetic moment and high magnetic ordering, but ferromagnetism was only observed below 10 K. In the same year, they prepared another ferromagnet by doping the ion of ClO_4^- into poly(3-methylthiophene) (PMTH)⁵⁷. The remnant magnetization of ClO_4^- -doped PMTH was $\sim 8.17 \times 10^{-4}$ emu/g, and the coercive field was 100 Oe. Moreover, weak ferromagnetic behavior was also observed in ClO_4^- partially doped poly(3-methylthiophene) (P3MT), as shown in Fig. 2⁵⁸. In this ferromagnet, polarons could be formed because of the ClO_4^- doping-generated charges, and weak ferromagnetism was introduced due to polaron-dopant interactions. The remnant magnetization of ClO_4^- -doped P3MT was $\sim 5 \times 10^{-3}$ emu/g, and the coercive field was approximately 130 Oe at 5 K (Fig. 2). At 300 K, the remnant magnetization decreased to 1×10^{-3} emu/g, and the coercive field was ~ 90 Oe.

In 2014, ferromagnetic ClO_4^- -doped poly(3-hexylthiophene) (P3HT) was prepared by the Rajca group⁵⁹. The unpaired electrons generated by the charge transfer between P3HT and ClO_4^- led to an open-cell structure that generated ferromagnetism at room temperature. When the storage time of ClO_4^- -doped P3HT reached 70 days, both the magnetization and coercivity almost disappeared. P3HT is a common electron donor polymer for organic solar cells^{60,61}.

In addition to charge transfer between P3HT and ClO_4^- , fullerene (C_{60}) and its derivatives are effective acceptors to combine with P3HT to form charge-transfer complexes. Recently, Ren et al.⁶² prepared a bulk heterojunction ferromagnetic film composed of P3HT nanowires and C_{60} . The saturation magnetization of the P3HT nanowire: C_{60} composite film measured in the dark was ~ 10 emu/cm³, and the coercive field was ~ 500 Oe. The saturation magnetization of the sample at room temperature increased when the sample was illuminated by a laser. It should be noted that a pure P3HT nanowire or C_{60} is nonferromagnetic. The ferromagnetism of the P3HT nanowire: C_{60} complexes may be attributed to the charge transfer between the P3HT nanowires and C_{60} . Moreover, amorphous P3HT: C_{60} also did not exhibit ferromagnetism, which suggested that the origin of the ferromagnetism in P3HT nanowire: C_{60} was closely related to the structure ordering. In 2016, Xie et al. built theoretical models to reveal that the mechanism of ferromagnetic ordering in P3HT nanowire: C_{60} was (i) charge transfer being able to break the closed-shell structure, leading to an open-shell structure, and (ii) the periodic crystal structure providing a long-distance spin ordering^{63,64}. In addition, the molecular configuration within the P3HT and C_{60} complex was also the key factor for the origin of room-temperature ferromagnetism. In 2019, according to theoretical simulations, spin polarization is generated in C_{60} when the pentagon of thiophene faces the pentagon of C_{60} . The spin polarization density disappears when the pentagon of

Fig. 3 The twisted TCNQ unit and magnetic hysteresis of organic ferromagnet p-TCNQ. **a** Twisted TCNQ unit with the spin density in the structure (indicated by the green dashed circle) showing the creation of spins on the connecting carbon atoms between the triazine rings and phenyl rings. **b** Magnetic hysteresis of organic ferromagnet p-TCNQ. Magnetization as a function of the applied field (H), recorded at 5 and 300 K. Measurements were performed in static field mode. The linear diamagnetic background was subtracted from all data. Spontaneous magnetization was clearly observed, even at 300 K. The internal hysteresis loops are shown in the inset. Reproduced from Mahmood et al.⁶⁶ with permission from Cell Press.

thiophene faces the hexagon of C_{60} ⁶⁵. For organic charge-transfer ferromagnets, at least four key factors determine their performance: (1) crystallization of organic materials providing long-distance spin ordering; (2) charge transfer inducing closed-shell structure breaking; (3) the molecular configuration; and (4) different electron–lattice coupling coefficients of the donor and acceptor. We think that further studies are needed to uncover more key factors.

To further improve the performance of organic ferromagnetism, polymerization was used to prepare polymeric ferromagnets. Baek et al. fabricated a polymeric ferromagnet (self-polymerized TCNQ) with a π -conjugated network structure⁶⁶. They observed ferromagnetic ordering in self-polymerized TCNQ even above 400 K. The ferromagnetic ordering in self-polymerized TCNQ was due to the broken π -bonding near the triazine moieties that generated inter- and intramolecular exchange interactions. The estimated Curie temperature of this polymeric ferromagnet was 495 K, which is the highest Curie temperature among the reported organic ferromagnets. The ferromagnetic property of self-polymerized TCNQ exhibited excellent stability under normal laboratory conditions. The magnetic response could be observed even after 2 years. The twisted TCNQ unit with the spin density in the structure and the field-dependent magnetization are shown in Fig. 3.

In addition to ferromagnetic hydrocarbons, known pure carbon-based ferromagnets⁶⁷ have also been realized, including graphite after proton irradiation^{68,69}, carbon

nanotubes⁷⁰, C_{60} ^{71,72}, and graphene^{73,74}. For sp^2 carbon materials, the orbital hybridization of C atoms leads to the formation of delocalized π bonds, and intrinsic diamagnetism should be generated. In 2007, Mathew et al.⁷⁵ vaporized C_{60} onto a silicon substrate and irradiated the C_{60} film with a proton irradiation dose of $6 \times 10^{15} \text{ H}^+/\text{cm}^2$ (ref. ⁷⁶). The proton irradiation produced local defects that led to local magnetic moments. This result was further confirmed by Kyu Won Lee et al.⁷⁷. In addition to proton irradiation, laser irradiation⁷⁸ and hydrogenation^{79,80} of fullerenes could also generate defects to induce ferromagnetism.

For graphene^{81,82}, ferromagnetism could also be generated by defects^{74,83}, topological defects^{74,84,85}, or hydrogen atom absorption^{81,86–88}. At room temperature, the saturation magnetization of grid-shaped graphene was 0.04 emu/g, and the coercivity was 558 Oe⁸⁹. Theoretically, the room-temperature saturation magnetization should be 0.87 emu/g if all the carbon atoms have unpaired electrons^{90,91}. Therefore, this result indicated that the number of carbon atoms contributing to ferromagnetism in grid-like graphene was only 0.008%. For the ferromagnetic behavior, the localized electron states at the grain boundaries of highly oriented pyrolytic graphite (HOPG) could lead to spin polarization and overall ferromagnetism, which could be observed through scanning tunneling microscopy (Fig. 4). Recently, the introduction of various chemical functional groups⁹² into graphene was also attempted to enhance the ferromagnetic ordering,

Fig. 4 Structure information and magnetization measurements on HOPG. **a** Schematic models of 2D in-plane magnetized grain boundaries propagating through bulk HOPG. **b** STM image of a grain boundary on HOPG showing a 1D superlattice with a small periodicity $D = 1.4$ nm. In-plane **c, d** SQUID magnetization measurements on HOPG after subtraction of the diamagnetic signal at 5 K and 300 K. Reproduced from Cervenka et al.⁹¹ with permission from the Nature Publishing Group.

including via graphene hydrogenation⁸⁸ and by reducing graphene oxide⁹³. In 2019, the new carbon magnet poly (meta-aniline) (m-PANI) was synthesized by Goto et al.⁹⁴, where iodine doping plays an important role in ferromagnetic interactions.

Although many organic ferromagnets have been made, including polymeric ferromagnets, carbon-based ferromagnets, and organic charge-transfer ferromagnets, it is difficult to fabricate organic ferromagnets with high Curie temperatures and large saturation magnetization and coercive field. On the one hand, organic ferromagnets need molecules with unpaired free radicals to form crystals through self-assembly, where it is difficult to obtain a stable structure. On the other hand, the mechanism of the origin of organic ferromagnetism is still only partly understood, which prevents us from fabricating an organic ferromagnet with good performance.

Organic magnetoelectric materials

Magnetoelectric coupling produces many novel physical effects for potential applications. Therefore, magnetoelectric coupling materials have great potential applications in compact, fast, and low-power multifunctional electronic devices, including energy harvesting devices, microwave devices, magnetoelectric memory, and magnetic sensors. In the following, we will present the progress in organic ferromagnetic magnetoelectric coupling.

Organic charge-transfer magnetoelectric materials

Tetrathiafulvalene (TTF) can form charge-transfer complexes with other organic acceptors. For TTF-*p*-chloranil (TTF-CA), the multiferroic property was observed⁹⁵. Moreover, TTF-*p*-bromanil (TTF-BA), as another organic charge-transfer complex, also had the multiferroic property⁹⁶. When the temperature was below 53 K, TTF-BA exhibited ferromagnetism due to the interaction between the spin and lattice. In addition, the magnetoelectric coupling effect was observed in TTF-BA crystals. The ferroelectric polarization could be changed by applying an external magnetic field owing to the spin-Peierls transition. The magnetic-field dependence of the electric polarization of TTF-BA at different temperatures is shown in Fig. 5a. In addition to magnetically controllable ferroelectrics of organic charge-transfer complexes, the phenomenon of electric-dipole-driven magnetism was also observed in the organic multiferroic charge-transfer complex κ -(BEDT-TTF)₂Cu[N(CN)₂]Cl⁹⁷, where the tunability of the spin polarization by an electric field was observed. The ferromagnetism of κ -(BEDT-TTF)₂Cu[N(CN)₂]Cl was induced by the polarized spin of the holes within the dimers. Consequently, both ferromagnetism and ferroelectricity were observed in multiferroic κ -(BEDT-TTF)₂Cu[N(CN)₂]Cl (Fig. 5b).

In 2012, Ren et al.⁶² explored the magnetoelectric coupling effect in the P3HT nanowire:C₆₀ complexes at room

Fig. 5 Organic magnetoelectric coupling. **a** Magnetic-field dependence of the electric polarization ($P \parallel b$) under $H \perp b$ at various temperatures in TTF-BA. **b** Temperature dependence of the conductivity of $\kappa\text{-(ET)}_2\text{Cu[N(CN)}_2\text{]Cl}$. Reproduced from refs. ⁹⁶ and ⁹⁷ with permission from the Nature Publishing Group.

Fig. 6 External field dependence of magnetization. **a** Room-temperature magnetoelectric character of P3HT nanowire: C_{60} ; the magnetization could be switched by a very small voltage. The electric field of the film sample was created parallel to the normal of the film, whereas the magnetic field was applied perpendicular to it. The measurements were performed in the dark. **b** Room-temperature magnetoelastic character of P3HT nanowire: C_{60} ; the (dark) magnetization could be switched by very small negative and positive strains. The magnetoelastic response should be antisymmetric. Reproduced from Ren and Wutting⁶² with permission from Wiley-VCH.

temperature. In the P3HT nanowire: C_{60} charge-transfer ferromagnets, the magnetization could be affected by the charge density. Once an external electric field was applied, the density of charge-transfer states could be enhanced, increasing the charge density to further enhance the open-shell structure. Consequently, the overall magnetization increased. The P3HT nanowire: C_{60} complexes showed a magnetoelectric coefficient of 40 mV/(cm Oe), as shown in Fig. 6. In addition to the P3HT nanowire: C_{60} complex, Qin et al.⁹⁸ also explored the magnetoelectric coupling effect in charge-transfer polymeric multiferroics (P3HT:PCBM complexes), the single-walled carbon nanotube (SWCNT) and

C_{60} system, and the P3HT nanowire:Au nanocluster^{99,100}. To further increase the magnetoelectric coupling coefficient, ferroelectric poly(vinylidene fluoride-trifluoroethylene) [P(VDF-TrFE)] was involved. As shown in Fig. 7b, the magnetoelectric coupling effect could be enhanced by introducing P(VDF-TrFE) as a ferroelectric layer between the C_{60} -SWCNT film and the electrode. Under an external electric field, the aligned dipoles in the P(VDF-TrFE) layer could increase the effective electric field in the C_{60} -SWCNT layer to enhance the open-shell structure, increasing the overall magnetization. As a result, the magnetoelectric coupling of these ferromagnetic charge-transfer complexes was

Fig. 7 The scheme, electric-field dependence of magnetization and M-H loops of all carbon complexes. **a** Scheme of the nanocarbon device structure. **b** Tunability of the magnetization by an electric field with and without the ferroelectric P(VDF-TrFE) layer. ON (OFF) means that the applied electric field ($1.8 \times 10^5 \text{ Vcm}^{-1}$) was turned on (off). **c** M-H loops of C₆₀-SWCNT/P(VDF-TrFE) bilayer devices under compressive and tensile modes. Reproduced from Qin et al.⁹⁹ with permission from Wiley-VCH.

enhanced. Moreover, the magnetization of these charge-transfer systems could be well tuned by an external stress, as shown in Figs. 6b and 7c.

Organic ferroelectric/ferromagnetic heterojunctions

Another effective way to realize the organic magnetoelectric effect is to fabricate ferroelectric/ferromagnetic heterojunctions. P(VDF-TrFE) is commonly used as a ferroelectric material. By using P(VDF-TrFE) and Terfenol-D as the ferroelectric and ferromagnetic layers, respectively, in 2001, Nan et al.¹⁰¹ prepared organic/inorganic flexible magnetoelectric films to well realize pronounced magnetoelectric coupling. Moreover, magnetoelectric coupling could also be observed by introducing ferromagnetic nanoparticles into ferroelectric films. P(VDF-TrFE) and Fe₃O₄ nanoparticles were used to prepare Fe₃O₄/P(VDF-TrFE) nanocomposites, where the

magnetoelectric effect was observed because of the elastic interaction between the Fe₃O₄ nanoparticles and macrodomains of semicrystalline P(VDF-TrFE)^{102,103}.

Multilayer structures with ferroelectric and ferromagnetic layers could effectively increase the magnetoelectric coupling effect^{104–106}. Ferromagnetic Terfenol-D and ferroelectric P(VDF-TrFE) were used to prepare multilayer magnetoelectric composites¹⁰⁷. The room-temperature magnetoelectric coupling coefficient of the multilayer P(VDF-TrFE)/Terfenol-D could reach 2 V/(cm Oe). Dong et al. prepared flexible magnetoelectric composites using Metglas and PVDF¹⁰⁸, where the magnetoelectric coupling coefficient reached as high as 7.2 V/(cm Oe).

Organic optomagnetic materials

As one of the most promising research areas of organic spintronics, optomagnetic coupling attracts our attention

because of the remarkable progress in understanding the interaction between light and spin. Optomagnetic coupling materials have great potential applications in faster and less dissipative magnetic random access memory and all-optical manipulation of the magnetization in magneto-optical recording. In the following, we will present recent developments in organic optomagnetic materials.

Tunability of magnetization by light

As shown in Fig. 8, the maximum magnetization of P3HT nanowire:C₆₀ was ~10 emu/cm³ in a dark environment. When P3HT nanowire:C₆₀ was excited by a 615-nm laser, the magnetization magnitude increased to 30 emu/cm³. Under external laser illumination, more charge-transfer states could be generated in polythiophene-C₆₀ crystals to further break the closed-shell structure, enhancing the overall magnetization.

Fig. 10 CPL spectra and magnetic field dependence of CPL. **a** CPL spectra of R/S-BN-CF, R/S-BN-CCB, R/S-BN-DBC, and R/S-BN-AF in the neat film state. **b** CPL (top) and fluorescence (bottom) spectra. **c** Circularly polarized light emission of chiral helical P3HT nanowires, where $g_{lum} = 2(I_L - I_R)/(I_L + I_R)$, I is the emission intensity, and σ^+ (σ^-) denotes right-handed (left-handed) circularly polarized light emission. **d** Magnetic-field-dependent spectra of g_{lum} . The inset shows magnetic-field-dependent g_{lum} values at 690 nm. Reproduced from refs. ¹¹⁸ and ¹²⁰ with permission from Wiley-VCH and Liang et al. ¹¹⁹ with permission from the American Chemical Society.

Thus, external light could tune the magnitude of the magnetization, presenting optomagnetic coupling in pure organic ferromagnets.

Although changing the intensity of the external light could alter the magnitude of the magnetization, presenting coupling between light and spin, changing the polarization state of light (with identical intensity) had little effect on the tunability of the magnetization of P3HT nanowire: C_{60} . Thus, new organic ferromagnets should be explored. Because the chirality and spin are closely coupled, many research groups have proven that chiral organic molecules could induce spin selectivity¹⁰⁹. As shown in Fig. 9a, when electrons are transported through chiral molecules, the transmission of electrons is strongly dependent on the spin orientation^{110–114}. Thus, fabricating organic chiral ferromagnets will provide a broad platform for further studies on optomagnetic coupling.

Compared to achiral organic ferromagnetic materials, the orbital angular momentum in chiral organic materials could be enhanced by the chirality^{115,116}. The orbital angular momentum enhances the spin-photon coupling effect in organic chiral materials^{65,117}. Qin et al. combined chiral P3HT nanowires with C_{60} to form chiral ferromagnetic P3HT: C_{60} complexes, where both the ferromagnetism

and magnetoelectric coupling effect could be tuned by polarized light. The magnetization of the chiral P3HT nanowire: C_{60} complexes was larger than that of the achiral P3HT nanowire: C_{60} complexes under light illumination with identical intensity. To further confirm that the chirality-induced orbital angular momentum was the main reason for the optomagnetic coupling, another chiral helical charge-transfer ferromagnet was prepared, where similar optomagnetic phenomena were observed¹¹⁷.

Magnetic-field-dependent polarization state of light

Due to the chirality-induced orbital angular momentum, circularly polarized emission could be observed in organic chiral materials (Fig. 10a, b)^{118,119}. The chirality-generated orbital angular momentum could lead to splitting of the lowest unoccupied molecular orbital (LUMO) and a significant decrease in the spin relaxation time. Therefore, the spin relaxation time is comparable to the recombination time in organic chiral ferromagnetic materials. The excited carriers recombine again after spin relaxation, leading to detectable circularly polarized light emission. As shown in Fig. 10c, d, the circularly polarized emission in organic chiral ferromagnetic materials could be tuned by an external

Fig. 11 CD spectra and transmissivity of chiral TPPS crystals. **a** CD spectra of right-handed and left-handed chiral TPPS crystal assemblies formulated with TPPS:DAC ratios of 1:2 and 1:4. **b** Magnetic-field-dependent transmissivity (T) of left/right-handed circular polarization components. The incident light was a linearly polarized laser of 360 nm. $\sigma+$ ($\sigma-$) denotes the transmission light of the right-handed (left-handed) circularly polarized component. Reproduced from Gao et al.¹²¹ with permission from the American Physical Society.

magnetic field¹²⁰. Under the stimulus of an external magnetic field, the spin polarization was enhanced in chiral ferromagnets, where the chirality-induced orbital angular momentum was further changed through the spin-orbit coupling (SOC). Because the orbital angular momentum determined the polarization state of the emission, circularly polarized emission was tuned by an external magnetic field, presenting the coupling between the polarized light and spin (Fig. 10d). Furthermore, because of the chirality-generated orbital angular momentum, both the transmissivity (T) of left (right)-handed circular polarization and circular dichroism (CD) could be tuned by an external magnetic field (Fig. 11)¹²¹. Moreover, molecular magnets with chiral ligands also presented chirality-dependent transmission^{122–124}.

Conclusion

In summary, the progress in organic ferromagnetic magnetoelectric and optomagnetic couplings is presented in this review. By designing novel structures, substantial progress has been realized for organic magnetoelectric and optomagnetic couplings. In organic charge-transfer complexes, magnetoelectric coupling effects can be realized through the spin ordering-driven electric polarization or the ferroelectricity driven by the spin ordering. The orbital angular momentum acts as an effective medium that determines the interaction between the photon and spin. Consequently, the chirality-generated orbital angular momentum plays a key role in the optomagnetic coupling effects in organic ferromagnets. Overall, organic ferromagnetic magnetoelectric and optomagnetic couplings lay solid foundations for mechanism studies and potential industrial applications. However, it should be noted that there are still many

unknown physical problems in this field, which need to be further studied.

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Conflict of interest

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