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Review

Modern History of Organic Conductors: An Overview

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Abstract: This short review article provides the reader with a summary of the history of organic conductors. To retain a neutral and objective point of view regarding the history, background, novelty, and details of each research subject within this field, a thousand references have been cited with full titles and arranged in chronological order. Among the research conducted over ~70 years, topics from the last two decades are discussed in more detail than the rest. Unlike other papers in this issue, this review will help readers to understand the origin of each topic within the field of organic conductors and how they have evolved. Due to the advancements achieved over these 70 years, the field is nearing new horizons. As history is often a reflection of the future, this review is expected to show the future directions of this research field.

Keywords: π–d system; Mott insulator; strongly correlated electron system; multiferroic; dielectric; photoconductor; Dirac electron system; single-component molecular conductor

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1. Introduction

At the beginning of this Special Issue of "Organic Conductors", we will briefly review the history of the organic-conductor research field (~70 years), during which tens of thousands of related papers have been published for readers to better understand the background and significance of the work collected in this issue. Note that the selected topics and references in this review are not exhaustive and that our purpose does not lie in comprehensively discussing each topic. Instead, we will limit ourselves to an overview of the course of the history, namely, how the studies concerning a specific topic were conducted and how they have been advanced. Although there still remains a number of important contributions and topics that are referred to in this review, many original papers, reviews, and books are cited, including those of closely related fields. Unlike the conventional manner of citing scientific literature, the references in this review were collected in a broad context and are listed in chronological order, rather than classifying them into particular topics. This was carried out to provide objective descriptions of the evolution and background of the various research topics in this field and the contributions of individual groups. Despite numerous references being cited, the reader can easily find the desired reference owing to their titles being included and their listing in chronological order. It is also expected that such a manner of citation would provide a comprehensive (underlying) connection between different studies, papers, and topics, i.e., how they have evolved by interacting with each other and different research fields. The reader can delve into the continuous, worldwide endeavors toward advancing the field of organic-conductor research by reading through the titles of the cited references, consequently developing an understanding of the field. After a brief summary of the older history of this field, we will provide an overview of the more recent progress that has been made in this field and the new trends that have developed in the last two decades. There are a number of other comprehensive reviews and books on the earlier stages of this field [1–21].

Constructing semiconducting materials comprised of organic compounds was first attempted by Akamatu, Inokuchi, and Matsunaga in 1954 [22], which has been recog-

nized as the emergence of a new research field, i.e., "organic (semi)conductors." Although the first "organic conductor" was unstable in air and possessed an unknown structure, it impacted the scientific community so greatly that it was succeeded by a series of significant findings, including the first organic metallic conductor TTF-TCNQ (TTF = tetrathiafulvalene, TCNQ = tetracyanoquinodimethane) in 1973 [23–29], doped polyacetylene in 1977 [30] (awarded the Nobel Prize in Chemistry in 2000), superconducting TMTSF (TMTSF = tetramethyltetraselenafulvalene) in the 1980s [31–66], and BEDT-TTF (BEDT-TTF = bis(ethylenedithio)tetrathiafulvalene) salts throughout the 1980s and 1990s [67–163]. This was followed by the advent of doped-fullerene (fulleride) superconductors in 1991, which demonstrated transition temperatures (T_Cs) of ~18–30 K [164–177].

1.1. Renaissance of Organic Superconductors

This historical overview should begin with a brief summary of the "organic superconductor age", during which the field rapidly progressed and expanded. The period, which occurred from the 1980s to the 1990s, is characterized by the worldwide efforts to discover the first organic superconductor, as well as the discovery of organic superconductors that succeeded the first and exhibited higher T_Cs, which was due to the dimensions of their intermolecular interactions being enhanced (Figure 1) [178–255]. These intermolecular interactions, which are indispensable for electrical conduction, are based on the overlap integrals of adjacent molecular orbitals. These overlap integrals are expected to be enhanced by introducing highly polarizable atoms, such as chalcogen atoms, onto the periphery of π-conjugated molecules. Such a synthetic strategy also leads to the stabilization of radical ionic molecular species in the solid state of charge transfer (CT) complexes, and a great number of new π-conjugated molecules, including metal-complex derivatives, were synthesized in this period of the 1980s to the 1990s [178–298]. There were four major families of organic superconductors established in this period, except for the fulleride superconductors. In the order of their appearance, they are based on the TMTSF [31–66], BEDT-TTF [67–163], DMET (DMET = dimethyl(ethylenedithio)diselenadithiafulvalene) [179–184], and M(dmit)$_2$ (M = Ni, Pd, Pt, etc; dmit = dimercaptoisotrithione or 1,3-dithiol-2-thione-4,5-dithiolate) [256–298] molecules. To emphasize their metallic and superconducting properties, organic conductors were often called "organic metals and superconductors" as well as "synthetic metals." In addition to their metallic and superconducting properties, the enhancement in the dimensions of their intermolecular interactions completely changed the crystal and electronic structures of organic conductors; specifically, they wiped out the prevailed image that organic conductors possess one-dimensional (1D) columnar (stacking) structures comprising planar π-conjugated radical ionic molecular species. The diversity of molecular arrangements in BEDT-TTF salts, which usually take on two-dimensional (2D) arrangements of BEDT-TTF cation radical species, far exceeds that of the molecular arrangements in 1D-type salts, such as TTF and TMTSF salts. The crystal structure variety of a compound is illustrated by the Greek letters placed at the beginning of its chemical formula, such as α-BEDT-TTF$_2$X (X = anions). The structural and electronic features of BEDT-TTF salts provided rich and unique findings for areas of chemistry and physics, which established the research field of organic conductors. However, during this period (~1980–2000), we also learned that similar molecules did not always produce CT salts with similar structural and physical properties. As metal *d*-orbitals can mix with ligand π-orbitals, species such as M(dmit)$_2$ distinguish themselves with some of their chemical properties. First, they are easily oxidized to provide stable radical anions and sometimes neutral radical species. Second, they have much narrower band gaps (~1 eV) between their HOMOs and LUMOs (HOMO = the highest occupied molecular orbital, LUMO = the lowest unoccupied molecular orbital) than other components of molecular conductors. In other words, the physical properties of M(dmit)$_2$ species may be governed by their LUMO and/or HOMO, depending on their oxidation state, crystal structure, and the existing thermodynamic conditions. Owing to these features, M(dmit)$_2$ and related salts have led to many new

and unique topics such as HOMO–LUMO inversion [273,274,284,285], single-component conductors, and Dirac electronic systems; the latter two are discussed below.

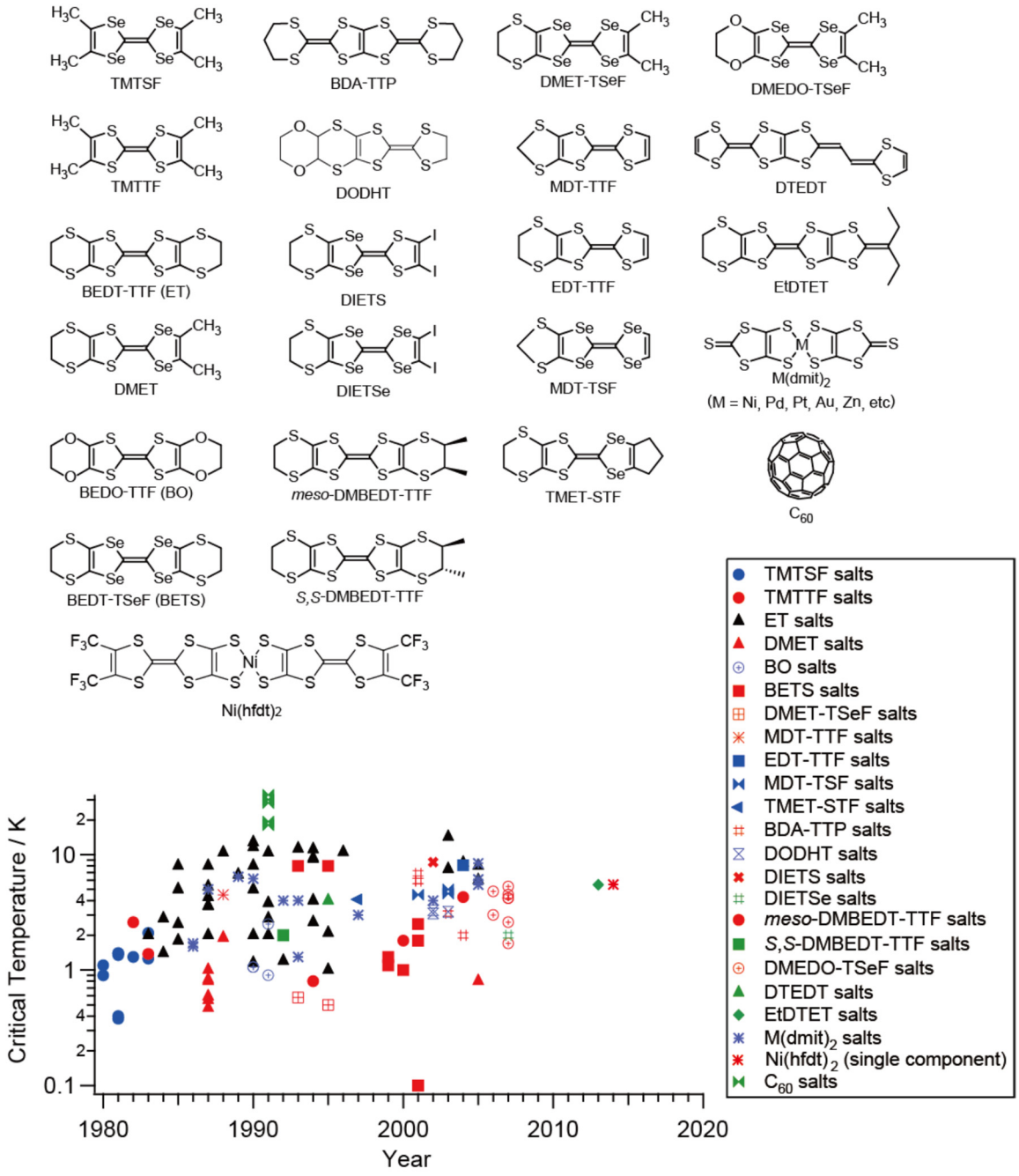


Figure 1. The history of the superconducting critical temperatures (T_Cs) of organic and related conductors (not an exhaustive list). Note that all the component molecules, except for $M(dmit)_2$ and C_{60}, belong to donor systems. Reproduced and modified from Ref. [21] with permission.

1.2. Beyond π-Systems: d-Electrons

1.2.1. The Rise of an Acceptor: DCNQI Salts

Toward the end of the "organic superconductor age", the merging of two extreme types of unpaired electrons, i.e., localized and delocalized electronic systems, was observed. The molecular conductors developed thus far in history (up until ~1985) were dominated by electron-donating molecular systems (donor systems), which are predominantly represented by the cation radical salts of TTF derivatives. This is natural when considering the

high polarizability and large van der Waals radii shared by chalcogen atoms, both of which being indispensable for the stability of the resultant radical species and the intermolecular interactions providing electrical conduction. Beginning with the first example of a purely organic metallic conductor, TTF-TCNQ [23–29], a strategy for developing more thermodynamically stable organic materials with enhanced conduction involved the prosperity of π-conjugated donor molecules possessing many chalcogen atoms at their peripheries. Meanwhile, electron-acceptor (acceptor) molecules, such as TCNQ, inherently contain highly electronegative (EN) atoms and electron-withdrawing functional groups. Such a molecular structure tends to attract and localize the mobile electrons on particular parts of the acceptor molecules, which appears to be disadvantageous for high conductivity. However, highly EN atoms and electron-withdrawing functional groups generally share another important chemical feature that is advantageous for intermolecular interactions, namely, strong coordination ability, which chalcogen atoms generally do not have.

In 1986, a series of highly conducting copper salts of organic acceptors attracted enormous attention because their electrical conduction was based on both the *d*-levels of the copper ions and the π-bands of the acceptors, i.e., π–*d* mixed-band conduction. The acceptors were quinone derivatives called DCNQIs (2,5-disubstituted *N*,*N*′-dicyanoquinonediimines) (Figure 2) [299–366]. They are distinguishable from the existing components in organic conductors because they can transform into stable radical anions and form covalent bonds with appropriate transition-metal cations, such as copper ions, to give CT complexes; this results in π–*d* mixed bands. Extensive studies on DCNQI salts revealed a number of unprecedented phenomena in the organic conductors they were a part of, including three-dimensional (3D) Fermi surfaces [302,331,334,341,346], metallic conductors with magnetic ordering [307,326,332,340,350], pressure-induced metal instability and reentrant metal–insulator transitions [301,303–305,309,310,326–328,337,360], dense Kondo effects [310,321,325,326,334], and charge/spin fluctuations [324,344]. Regarding such hybrid band structures, DCNQI salts contain characteristics of 1D and higher-dimensional conductors, where they exhibit one or both types of characteristics depending on their chemical compositions and the implemented thermodynamic conditions. This puzzling feature of DCNQI salts provided researchers with a deeper understanding of organic conductors, but more uncertainty also emerged. The studies conducted on organic conductors thus far have provided some current, important notions in related research fields, such as charge separation. This term originally referred to the deviation in the electron density of the conduction pathways, from evenly distributed, i.e., delocalized, to periodically localized patterns of metal–insulator (M–I) transitions. The term "charge separation" is more frequently used in the field of semiconductor devices such as field-effect transistors and solar cells [367–428] than in the field of organic crystalline conductors. During ~1985–2000, ideal samples and elaborate techniques for studies on the complicated behavior of DCNQI salts were timely provided one after another; for example, studies regarding alloys [309,313,325,326,333,355,357] and selectively deuterated [328–333,340,343,345,354] DCNQI derivatives and fine-tuning of delicate pressures [303–305]. In addition to these researchers' endeavors, other successful studies have provided advanced techniques for crystalline molecular conductors to exhibit novel and unique conducting properties; namely, they were soft and thus highly sensitive to perturbations, such as applied pressure or magnetic fields. These properties make these crystalline molecular conductors superior to conductors composed of harder materials, as the latter do not exhibit responses to such perturbations as clearly as the former do. The key to success was also based on a close and efficient collaboration between chemists and physicists, which has always been present at one time or another in this field. In this way, the "π–*d* interaction" became a popular research topic and a desirable research target in the field of organic conductors. This research trend was followed by π–*d* systems implemented in donor-based conductors [429–518], as shown in the next subsection.

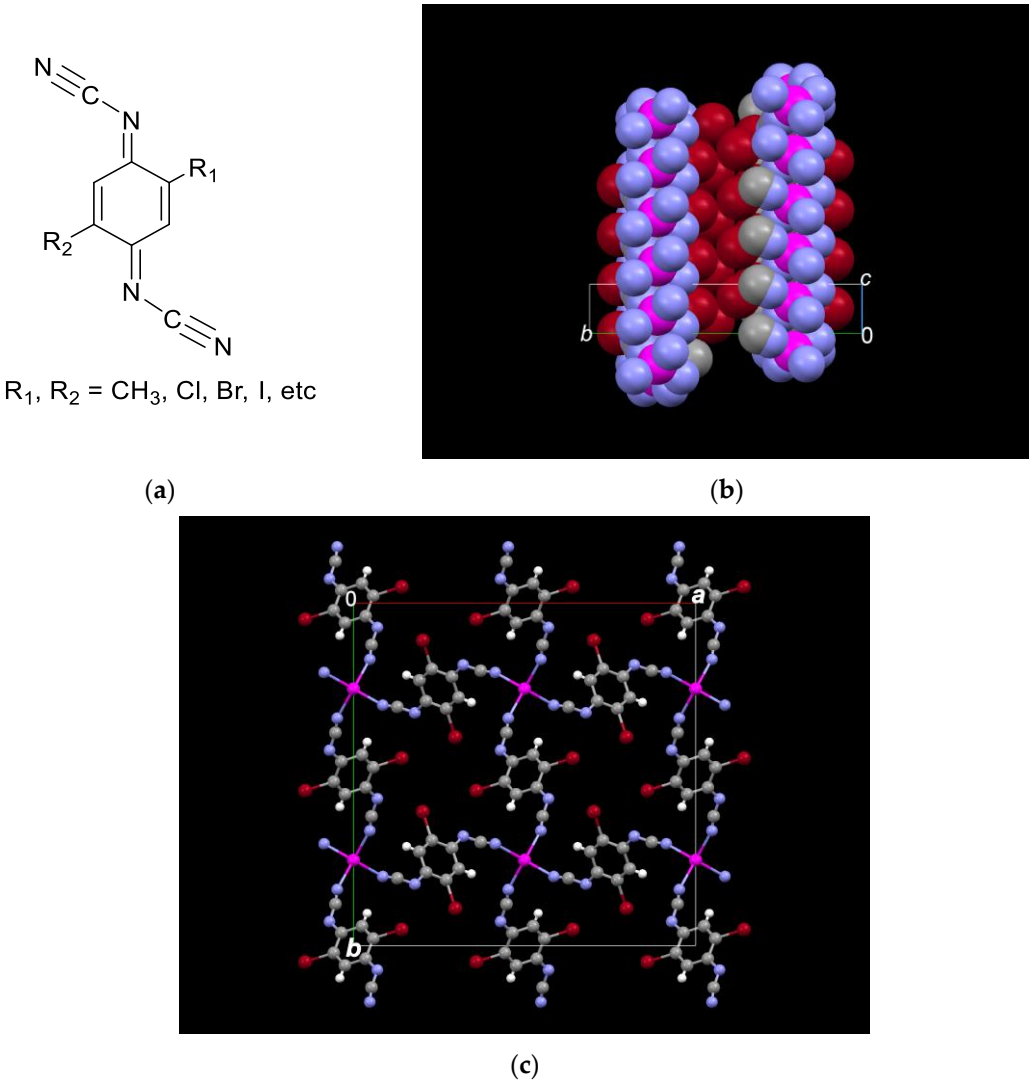
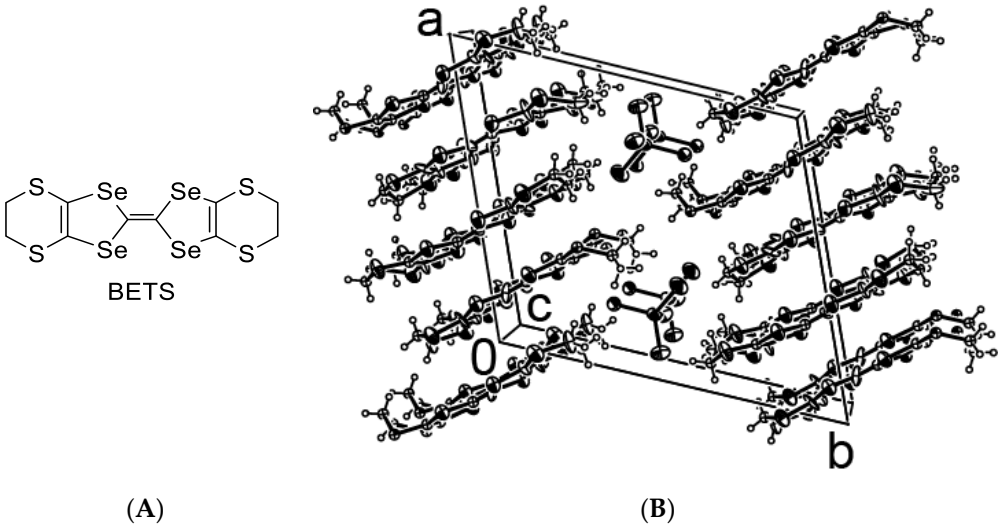


Figure 2. (**a**) The structure of the DCQNI (DCQNI = 2,5-disubstituted *N*,*N*′-dicyanoquionediimine) molecule; (**b**,**c**) A typical crystal structure of a Cu salt: Cu(Br$_2$-DCNQI)$_2$. The white, grey, purple, pink, and dark-red spheres designate H, C, N, Cu, and Br atoms, respectively. Note that every –NCN group at the end of DCNQI makes a coordination bond with a Cu ion in a tetrahedral geometry. Reproduced and modified from Refs. [519,520] with permission.

1.2.2. Comeback of the Donor Dynasty: BETS Salts with Magnetic Anions

In parallel with the extensive examination of DCNQI salts, many researchers have attempted to synthesize a similar system based on donor molecules since ~1990. The prominent examples of unique conductors based on π–*d* interactions are a series of CT salts of a BEDT-TTF derivative called BETS (bis(ethylenedithio)-tetraselenafulvalene) [451–518]. Various derivatives of BEDT-TTF have been extensively explored from an early stage of the field [451–462]. BETS was first discovered in 1983 [454]; however, it was not until 1993 that BETS was suddenly paid significant attention when λ-(BETS)$_2$GaCl$_4$ was found to be a new superconductor with nearly the highest T_C (~10 K) among those of the organic compounds known at the time [463]. Almost at the same time, λ-(BETS)$_2$FeCl$_4$ (Figure 3), an isostructural salt of λ-(BETS)$_2$GaCl$_4$, was reported [464] and garnered more attention than the preceding non-magnetic salt λ-(BETS)$_2$GaCl$_4$. This was because λ-(BETS)$_2$FeCl$_4$ and succeeding, related salts united the two main streams of organic superconductors and organic π–*d* systems. For example, some BETS salts, κ-BETS$_2$FeX$_4$ and λ-BETS$_2$FeX$_4$ (X = halogen atoms), have been able to achieve what DCNQI salts could not; examples include (high magnetic) field-induced superconductivity [465,481–483,485,486,489,490,494,497,500,501,504] and superconductors with magnetic ordering in their ground

states [471–475,477,479,480,497,498,504]. The 2D conduction sheets in λ-$BETS_2FeX_4$ serve as ideal samples for studies on the Fulde–Ferrell–Larkin–Ovchinnikov (FLLO) state, a theoretically predicted superconducting state where the order parameter oscillates in real space [490,501,513,514,521–524]. The common key feature in these π–*d* systems is weak or moderate π–*d* interactions, without which the systems could not exhibit unusual behavior based on both localized (*d*-electrons as spins) and delocalized (π-electrons as carriers) electronic characteristics. However, these interactions are far more difficult to achieve than originally assumed. A great number of TTF-type donor salts with paramagnetic metal complex anions were synthesized and their electrical and magnetic properties were measured [429–450,464–518]. Similarly, a large number of TTF-type molecules and their cation radical salts bearing stable radical moieties, such as nitroxide derivatives, were synthesized and were examined. However, most of them turned out to be antiferromagnetic semiconductors or even diamagnetic insulators in the ground state, except for a limited number of successful examples [525–528]. It is substantially difficult to finely tune the interactions between localized and delocalized unpaired electrons for them to coexist. Since then, researchers have begun paying more attention to "crystal designing" than "molecular designing", the former being a trend that also emerged in related research fields such as supramolecular chemistry [529–546] and those involving metal–organic frameworks (MOFs) [547,548].



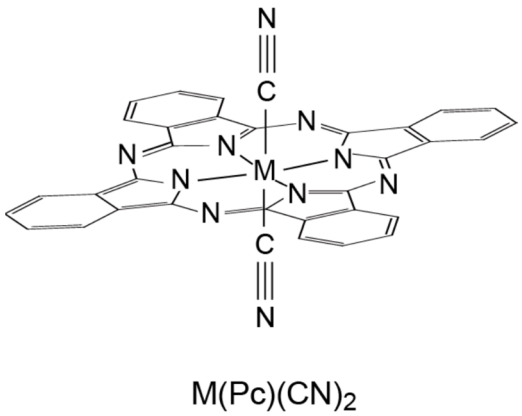
(**A**) (**B**)

Figure 3. (**A**) The chemical structure of the BETS (BETS = (bis(ethylenedithio)-tetraselenafulvalene)) molecule; (**B**) A typical crystal structure of the magnetic superconductors: λ-$(BETS)_2FeCl_4$. Note that one of the ethylene groups at the end of BETS is bent toward $FeCl_4^-$ to favor the π–*d* interaction. Reproduced from Ref. [511] with permission.

1.2.3. The Return of 1D Systems: Axially Ligated Iron(III) Phthalocyanines

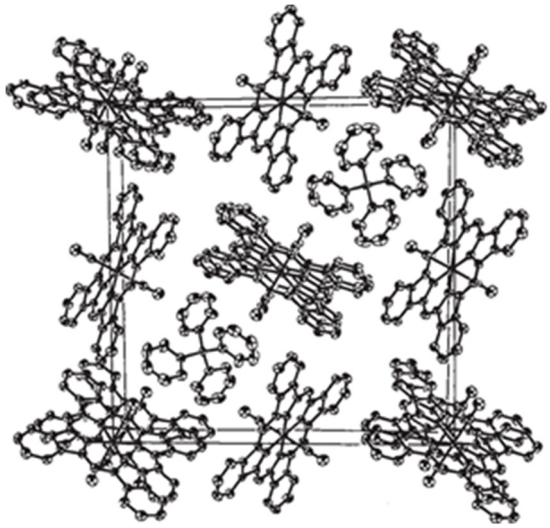
There is another unique π–*d* system, where both delocalized π-electrons and localized *d*-electrons are located on the same molecules, i.e., transition-metal phthalocyanines (MPcs). Owing to the superior stability and ready availability of MPc derivatives, there have been many pioneering works, including those in the 1980s that investigated (pseudo)halogen- or oxygen-ligated Pc polymers [549–552], single-component conductors [553], π–*d* interactions in Fe(III)Pc complexes [554], electrochemically synthesized $(NiPc)_2AsF_6$ complexes [555], and iodine-doped CuPc complexes [556] toward 1D molecular conductors. Then, in 1990, hybrid derivatives were discovered, i.e., electrochemically synthesized radical cation salts of axially ligated MPcs ($M(Pc)X_2$; M = metal cation, Pc = phthalocyanines and their derivatives, X = (pseudo)halogen monoanions as axial ligands; Figure 4) [557–578]. The studies on $M(Pc)X_2$ complexes also evaluated their crystal designs, as mentioned above, but most of them exhibited a similar columnar stacking structure with a 1D conduction band. The most intensively studied examples include $TPP[M(Pc)(CN)_2]_2$ (TPP = tetraphenyl phosphonium,

M = Co(III), Fe(III)) complexes. Because of the steric hindrance of the axial ligands, the Pc ligands stack with each other via limited π–π overlap (~single benzene rings). This produced narrow bands and semiconducting behavior in a wide variety of the $M(Pc)X_2$ salts, whether the metal centers were paramagnetic or not. Their slipped stacking pattern distinguishes $M(Pc)X_2$ salts from other organic conductors composed of planar molecules, which results in a number of unique and puzzling electrical properties in the former. Despite limited π–π interactions existing exclusively along the 1D columns, these salts exhibit high conductivity and no obvious transitions at ~2–300 K, which apparently contradicts the idea of 1D-metal instability (Peierls instability) that was established in the early stages of this field [579]. The commonly observed semiconducting behaviors of $M(Pc)X_2$ salts are not of a thermally activated type, and their reflectance spectra show a Drude-type dispersion [558,559]. This semiconducting behavior suggests the presence of small energy gaps originating from strong electron correlations and/or other reasons. The ground state of a $Co(III)(Pc)X_2$ salt has been clarified as a charge-ordered (CO) state [568], which is the typical ground state of thermodynamically unstable 1D conductors. The CO state has been acknowledged since the early stages of this field and has been gaining increasing attention due to its connection with superconductivity [580–625]. The electrical behavior of complexes bearing paramagnetic metal ions, such as Fe(III), is more unusual than that of complexes bearing diamagnetic metals. Furthermore, π–*d* interactions manifest under giant negative magnetoresistance (GNMR), which is clearly observed generally at $T \leq 50$ K and $H \geq 10$ T and depend on the magnetic-field direction relative to the stacking axes [557–578]. Unlike BETS and DCNQI salts, $M(Pc)X_2$ salts apparently belong to 1D systems, which are known to be inherently unfavorable for metallic conduction at low temperatures [579]. However, GNMR is generally observed at low temperatures; thus, metal-like high conduction is required at low temperatures. Therefore, the observation of GNMR in such a series of 1D systems is surprising. Accordingly, these π–*d* systems have demonstrated that organic conductors are a sufficient choice for studying solid-state physics on a much wider scale than was originally possible regarding cooperative phenomena.



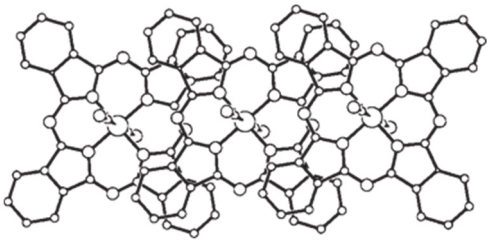
$M(Pc)(CN)_2$

(**a**)



(**b**)

Figure 4. *Cont.*



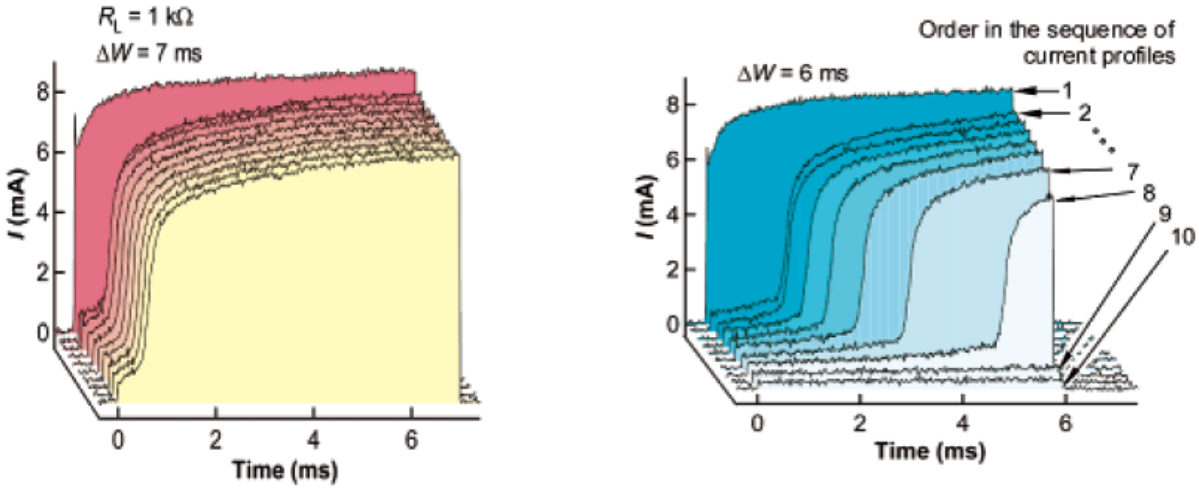
(**c**)

Figure 4. (**a**) The molecular structure of a metal phthalocyanine (MPc) $[M(Pc)(CN)_2]^n$ unit ($0 \leq n \leq 1$); (**b**) A typical crystal structure of $Cat[M(Pc)X_2]_2$ salts (Cat = onium monocations): $(C_6H_5)_4P[Fe(Pc)(CN)_2]_2$; (**c**) A close-up view of a 1D column of $[Fe(Pc)(CN)_2]^{0.5-}$. Reproduced from Ref. [559] with permission from The Royal Society of Chemistry.

1.3. To Be or Not to Be a Conductor—That Is the Problem: Fluctuation

Researchers that were active during the "organic superconductor age" had gradually realized that the enhancement of superconducting (SC) T_Cs could also provide an understanding of the insulating and/or magnetic phases of neighboring SC phases. In fact, it has often been pointed out by both theoretical and experimental studies that the fluctuation in charge and/or spin degrees of freedom could play an indispensable role in a potential, the universal mechanism behind SC transitions in organic and inorganic compounds [626–646]. As a result, the strategy for developing organic superconductors with higher T_Cs drastically changed from involving the stabilization of metallic states to employing the destabilization of metallic states [647–655]. In pursuing this strategy, synthetic efforts towards new conductors, even those where insulators had resulted instead, led to successive findings of unprecedented physical properties and various phase transitions [228,241]. All of these features originate from various types of degrees of freedom that are characteristic of molecular crystals, i.e., "molecular degrees of freedom." The variety of these degrees of freedom is important, in addition to their comparable thermodynamic stabilities and their states being incompatible with each other. These properties have been observed in various forms, such as fluctuation [626–646], hidden states [42,647], and field-induced cascade transitions [40,492].

As studies on organic conductors have progressed, so have the experimental tools/techniques used to observe electronic behavior; namely, rapid progress has been made in both the variety and specification of these tools, which has provided us with high magnetic fields [656–682]; high time- [683–710] (Figure 5), space-, and/or energy-resolutions [365, 711–721]; and high (hydrostatic or uniaxial) pressures [722–731]. As a result, the term "organic conductor" now includes a wide variety of compounds and components, such as organic polymers, inorganic ions, metal complexes, metal clusters, and organic molecules possessing electrical properties ranging from those of insulators to those of superconductors (depending on the circumstances). Thus, organic conductors are now better described as "molecular materials", which can be defined as a unique group of compounds with well-defined chemical formulae and crystal structures that are used in studies on cooperative phenomena.



(**a**) (**b**)

Figure 5. Sequential transient photocurrent measurements under synchronized voltage and photoirradiation pulses using the single crystal of α-(BEDT-TTF)$_2$I$_3$ and a four-probe method at 115 K, where the material is in an insulating phase. Photoirradiation was synchronized only at the first voltage pulse (11 V) and was turned off for the second and following pulses: the voltage pulse widths (Δ*W*) are (**a**) 7 ms and (**b**) 6 ms. Note that Δ*W* drastically affects the current, i.e., the relaxation time of the highly conducting state (HS) after the cessation of photoirradiation, while Δ*W* does not affect the resistivity in the HS. Adapted from Ref. [693] with permission.

Besides the basic studies on crystalline organic conductors, organic CT salts, and related compounds, semiconducting thin films in field-effect transistors (FET), batteries, luminescence devices, and non-linear optics have also been extensively studied since the 1970s [367–428]. Studies on such "organic devices" have become well established in various research fields of both academia and industry. Since there are a great number of reviews and books detailing such studies [382–385,397,398,401,402,417–428], we will not go into detail here.

Overall, the history of "organic conductors" involved the discovery of new, surprising materials and investigations into their unique properties. Selected landmark studies from the last two decades will be described in the following sections.

2. Recent Progress and New Trends

The field of organic conductor research has made steady progress over the years, beginning with organic semiconductors based on aromatic hydrocarbons and evolving to include organic metals, organic superconductors, and organic magnetic (super)conductors. Alongside such progress, the field has also evolved to include a broader range of topics. Advancements in the field have been achieved by incorporating heteroatoms, mainly chalcogen atoms and metal ions, in the aromatic hydrocarbons responsible for electrical conduction. Consequently, the component molecules of conductors become so diverse that they should be referred to as molecular conductors/materials rather than organic conductors. At the same time, experimental and theoretical tools used to study these types of materials have also made remarkable progress in recent decades. These new materials and new methods have accelerated the advancement of one another, which led the field in new directions. For example, since approximately the "organic superconductor age", particular types of organic insulators have been intensively studied for various reasons.

2.1. Mott Insulators: Mysterious Clues to Superconductors

Mott insulators [732–749], which are often closely related to high-T_C superconductors of both organic (Figure 6) and inorganic nature, have been paid particular attention because they are considered key compounds for clarifying a possibly universal high-T_C mechanism. Mott insulators are characterized by a paramagnetic half-filled band. Although

such an electronic feature appears to be that of metals, the unpaired electrons cannot travel through these solids because of the strong electron correlation. As a result, they have energy gaps called Mott (Mott–Hubbard) gaps at the Fermi level, which makes them different from common insulators, i.e., band insulators. Band insulators are diamagnetic, so they do not have unpaired electrons. By applying the band theory, Mott and band insulators could correspond to high and low spin states of a given band structure and band filling, whose differences originate from the on-site Coulomb energy. However, Mott insulators are intrinsically beyond the band theory, as their electron correlation is due to many-body problems instead of Coulomb repulsion between a pair of electrons. Currently, many organic and inorganic compounds are known as Mott insulators. Owing to their unique insulating mechanisms and potential to serve as high-T_C organic superconductors, many researchers have attempted to control the conducting properties of such compounds using chemical ("chemical pressure") and/or physical (applied high pressure) methods [649–655,722–731]. This is generally referred to as bandwidth control. Band-filling control, which is typically performed by synthesizing mixed crystals (doping), is frequently attempted with inorganic compounds, while bandwidth control is rather typical of organic conductors owing to their soft structures. Meanwhile, except for limited kinds of salts [196,309,313,325,326,333,355,357], the doping of crystalline CT salts is rather difficult [750]; the mixing of isostructural compounds with different electron counts often resulted in unexpected, pure compounds with different crystal structures or, more frequently, failed to yield well-defined crystalline solids. To solve this problem, a new method using photochemical reactions has been established [21,519,520,751–759].

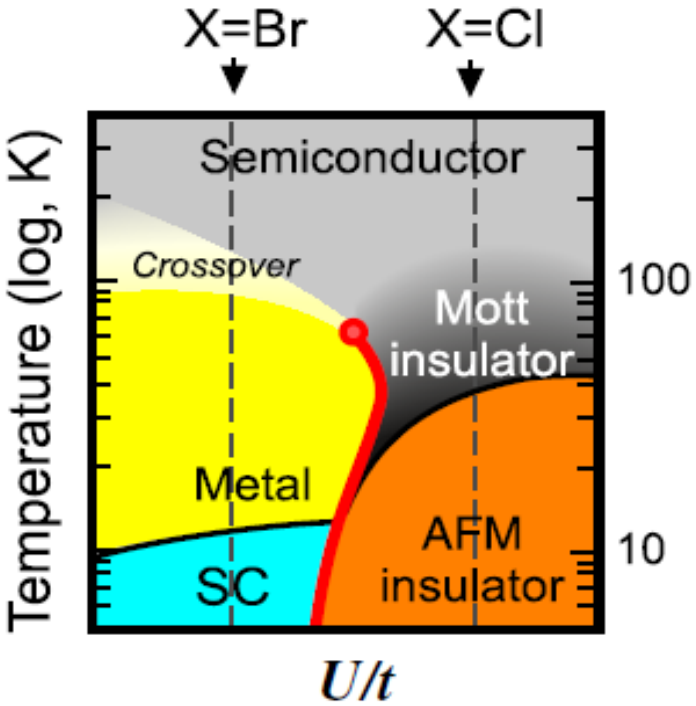


Figure 6. A schematic phase diagram of κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]X (X = Br, Cl) complexes, which are considered typical, organic Mott insulators and superconductors depending on the anion X. An increase in the horizontal axis (U/t) corresponds to a decrease in the actual pressure. Adapted from Ref. [704] with permission.

Similar to SC transitions, various kinds of M–I transitions have been extensively studied. Naturally occurring metals, i.e., the elemental metals, do not exhibit M–I transitions, except for tin. The insulating phases are generally induced by order–disorder transitions in a part of the crystal structures, charge and/or magnetic orderings, as well as changes in the temperature/pressure. Because there can be different (meta)stable states

in organic conductors that exhibit nearly equal stabilities, and because they frequently have low-dimensional electronic structures, a fluctuation between different states [626–646] or even the disappearance of phase transitions [41,647,648] are often observed. Both are characteristic of organic conductors, in addition to the M–I transitions.

2.2. Between Electron and Lattice Systems: Proton Dynamics

As protons are most frequently included in organic compounds, they have been paid significant attention in attempts to control the electron dynamics and resultant physical properties of organic conductors [535,760–772]. A new series of dielectrics [773–783] and multiferroics [784] based on organic CT salts have been synthesized/discovered. Multiferroics are materials that combine multiple order parameters, such as ferroelectrics and ferromagnets, where the spontaneous ordering in the magnetic and dipole moments simultaneously occurs within the same temperature range. Generally, magnetic orderings are observed at low temperatures, while the orderings of electric dipoles are observed at high temperatures. Thus, their coexistence in a single organic or inorganic material at the same temperature seldom occurs. Although their performance and temperature ranges have yet to be improved for practical applications, they have been employed in representative and successful studies derived from organic conductors, where various phenomena often couple together. In relation to proton dynamics, organic proton conductors [782] and organic conductors bearing components that participate in hydrogen bonding have also been developed [770,771]. Regarding the latter, it is extremely difficult to make the conduction π-electrons and the protons of hydrogen bonds interact with each other, even if they are located on the same molecules. This is likely due to the differences in their energy scales. Such subtle material design requires the extremely fine-tuning of energy levels and crystal structures, similar to the π–*d* systems. In fact, an analysis of non-deuterated and deuterated samples revealed that deuteration qualitatively affects conduction behavior [771] (Figure 7), which was also observed for DCNQI salts. The mechanism of this is currently under study. Regarding dielectrics, an electronic mechanism is often noted for some molecular CT salts instead of proton/ion displacement [785–795]. The dielectrics with an electronic mechanism comprise the insulating, i.e., CO, phases of organic conductors. Such dielectrics are characteristic of organic conductors, exemplifying the rich variety of their electronic states.

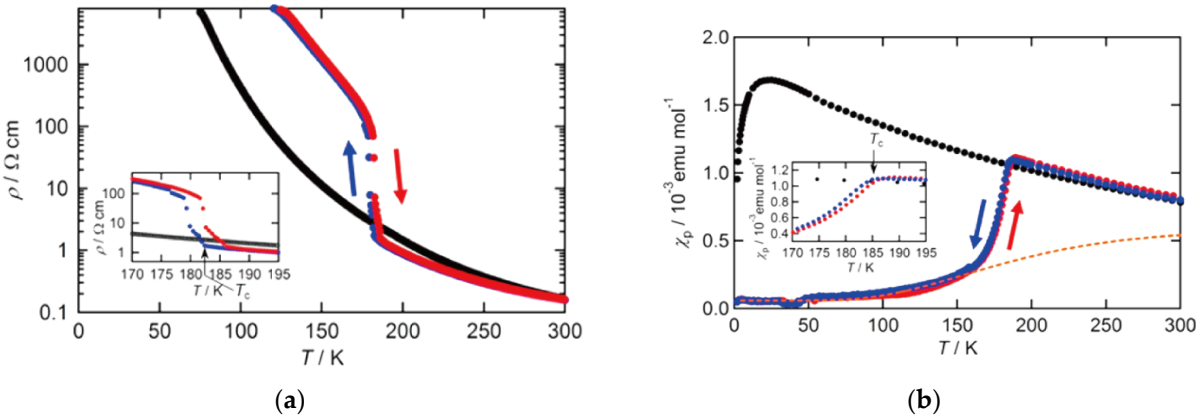


Figure 7. Temperature dependence of the physical properties of κ-H_3(Cat-EDT-TTF)$_2$ (κ-H) and κ-D_3(Cat-EDT-TTF)$_2$ (κ-D): (**a**) electrical resistivity measured using a single crystal; (**b**) magnetic susceptibility measured using a polycrystalline sample. In both figures, the blue and red circles denote the data observed in the cooling and heating processes, respectively, for κ-D, while the black circles denote the data for κ-H observed in the cooling processes. The orange broken curve in (**b**) represents the best fitting curve for the κ-D data using a singlet–triplet dimer model with an antiferromagnetic coupling of $2J/k_B \sim -600$ K. Adapted from Ref. [771] with permission.

2.3. Light Control: Unique Properties Otherwise Impossible

In addition, the extensive development and recent advancement of organic photoconductors and related materials are notable [796–826]. In contrast to the ground states of these materials, which most scientists in the field of the organic conductors have been interested in, their photoexcited states correspond to extremely high energy states that thermal excitation could not reach. Thus, photoirradiation may provide us with a new method for discovering new physical properties and structures. Therefore, instead of simply synthesizing photoconductors based on organic compounds, the development of these materials involved inducing novel physical properties and unique electronic phases by photoexcitation. This methodology was significant because many of these new properties had never been observed by simply controlling the temperature and/or pressure conditions. The development of these kinds of materials requires knowledge of organic conductors and the techniques used for their development, even though most of these materials are insulators/intrinsic semiconductors under dark conditions or without photoirradiation of appropriate wavelengths. Similar to photovoltaics and solar cells, the charge separation under photoirradiation and the lifetimes (relaxation times) of photocarriers produced thus far are crucial for ensuring the efficiency of photoconductors. Whether organic or inorganic, ordinary insulators typically exhibit an increase in conductivity of $\leq$20–30% at room temperature under ultraviolet-visible (UV-Vis) photoirradiation. Most of the increase in conductivity can be explained by heating effects involved with the irradiation, i.e., thermally activated carriers, and the net photoconductivity is generally ~2–3% of the original dark conductivity [21,520,759]. The relaxation of photoexcited carriers is generally quick, as long as the optical absorption occurs as resonance with UV-Vis (~10^{15} Hz), which is a typical energy range for HOMO–LUMO transitions in organic molecules used as conductors. Thus, a strategy for stabilizing photoexcited carriers is required, in addition to the design of conductors. In this sense, organic photoconductors can be regarded as an advanced design of organic conductors. Combining the typical component molecules of organic conductors, such as TTF derivatives, with well-known photosensitive molecules, such as bipyridyl derivatives, has resulted in various kinds of donor–acceptor type CT salts being reported for new organic photoconductors and related materials [796–826]. However, difficulties were encountered regarding the control of the donor–acceptor interactions toward producing a sufficient number of carriers, i.e., photoexcited electrons and holes, with sufficiently long relaxation times. Strong CT interactions lead to quick recombination between photoexcited electrons and holes, while weak CT interactions lead to an insufficient number of photoexcited electrons and holes. Additionally, if the photosensitive moieties are bulky, the formation of the conduction pathway is hindered. This is due to the close proximity of the π–π interactions between the main parts of the molecules that are responsible for conduction. Based on previous encounters with this kind of problem during the molecular and crystal design of π–*d* systems, several solutions have been proposed. One of the strategies involved insulating common organic CT salts with donor–acceptor mixed stacking structures, denoted as D_nA_m (n, m = 1, 2, 3, . . .) [520]. For example, if n = 1 and m = 2, the unit cell is rich in acceptor A and an infinite . . . A–A–A . . . network may form, based on both stacking and side-by-side overlaps serving as conduction pathways (Figure 8). This situation is more favorable if the molecular size of A is larger than that of D. The requirement for actual conduction is to produce carriers in the conduction pathways, which is often called "doping" and can be performed by the photoexcitation of the CT bands between acceptors and donors. If the excited states are stabilized by the CT interactions, similar to exciplexes [424,425,428,799,817], the relaxation times of the resultant carriers can be sufficiently long. Based on this idea, (para)magnetic photoconductors have been developed based on stable diamagnetic insulators, where strong interactions between photoexcited localized spins and photocarriers manifest in a Kondo effect (T_K ~ 100–120 K) [21,520,759,808]. Using the unusual stability of the photoexcited states of some molecular CT salts, a novel type of material for photon energy storage is under investigation [823,825].

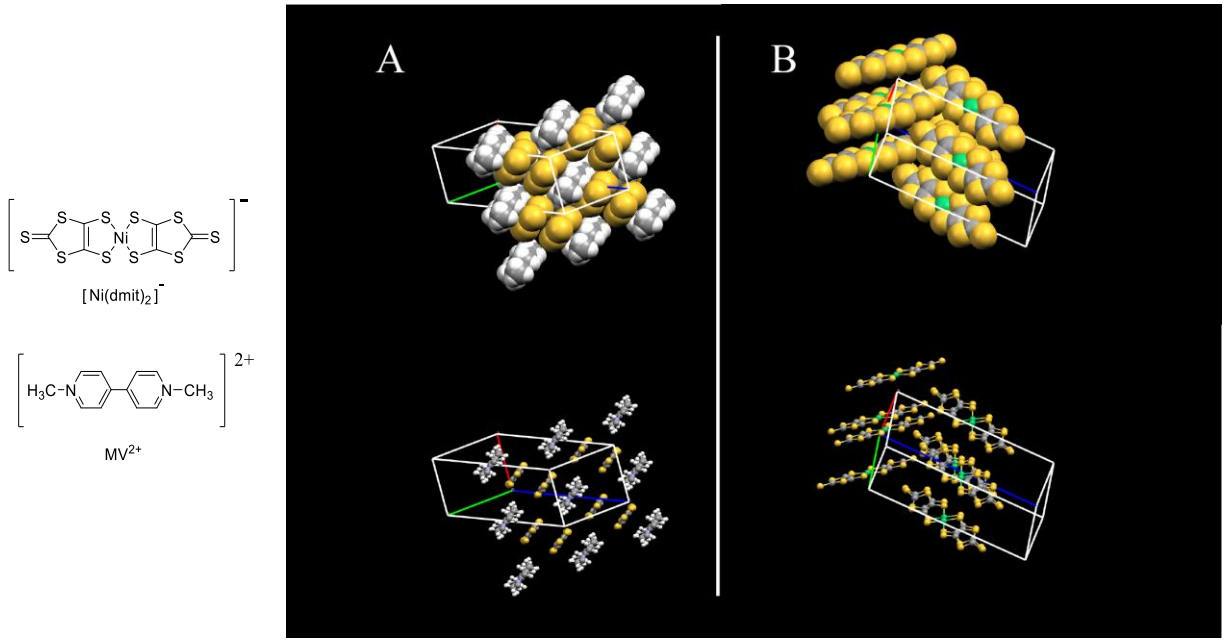
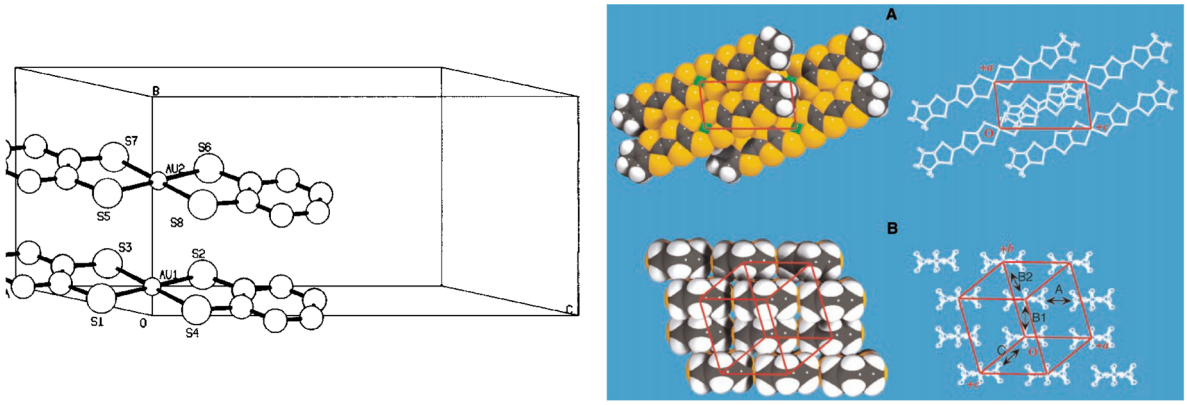


Figure 8. The crystal structure of a photomagnetic conductor, $MV[Ni(dmit)_2]_2$: (**a**) MV^{2+} and $[Ni(dmit)_2]^-$; (**b**) Molecular packing motif in van der Waals (upper) and ball-and-stick (lower) models; (**A**) Chemical structures of MV^{2+} and $[Ni(dmit)_2]^-$ and (**B**) 3D conduction pathways composed of $[Ni(dmit)_2]^-$. $MV[Ni(dmit)_2]_2$ is a diamagnetic insulator under dark conditions, but it turns into a metallic substance with localized spins on the MV^{2+} species under UV irradiation, thus exhibiting the Kondo effect at low temperature. The UV (~375 nm) irradiation triggers a CT transition between MV^{2+} and $[Ni(dmit)_2]^-$.

2.4. Single-Component Molecular Conductors: The Simplest and Most Difficult Molecular Conductors

Throughout the development of organic conductors, a molecular version of elemental metals, i.e., single-component molecular conductors (SCMC), has been a desirable target (Figure 9a) [729,827–882]. Unlike CT salts, SCMCs should have simpler crystal structures because they consist of a single type of molecular species. However, it turned out to be extremely difficult to produce such materials. Since the late 1980s, pioneering work towards these materials has been carried out by some independent groups. They obtained diamagnetic single crystals of neutral species [827–829] or paramagnetic and highly conducting/metallic polycrystalline samples of neutral species with unknown structures [830]. In 2001, i.e., nearly half a century since the beginning of this research field, the first SCMC with a well-defined crystal structure was developed based on a Ni–dithiolene complex molecule, $Ni(tmdt)_2$ (tmdt = trimethylenetetrathiafulvalenedithiolate) (Figure 9b) [831,832,838]. The SCMC exclusively contains neutral molecules of the same kind and produces carriers via the overlap between its HOMO and LUMO bands, similar to how semimetals produce carriers [837,873]. Because all the planar molecules are densely packed to ensure their equal contribution to conduction, the resultant conduction properties are 3D; however, there remains some anisotropy that is reflective of that of the component molecules. In principle, the crystal and electronic structures of SCMCs are uniform, and thus there should be usually no charge ordering or dimerization to make the material insulating. Consequently, most SCMCs are thermodynamically stable metals. However, SCMCs exhibiting an antiferromagnetic transition ($[Au(tmdt)_2]$) [843] and a coupled electric and magnetic transition ($[Cu(dmdt)_2]$) (dmdt = dimethyltetrathiafulvalenedithiolate) [857] are also known. Among the SCMCs, $[Ni(hfdt)_2]$ (hfdt = bis(trifluoromethyl)tetrathiafulvalenedithiolate) exhibits a superconducting transition (onset $T_C \leq 5.5$ K under 7.5–8.7 GPa) [729]. Recently, an increasing number of new SCMCs, most of which containing Au(III)–dithiolene complexes, have been reported [729,827–882]. Furthermore, new SCMC materials have been recently found to

possess unique electronic band structures called Dirac cones [869–872,875,876,878,880–882], which will be discussed in the next section.



(**a**) (**b**)

Figure 9. Crystal structures of single component molecular materials: (**a**) A 1D soft Mott insulator, $Au(bdt)_2$ (bdt = benzene-1,2-dithiolate); (**b**) A 3D metallic conductor, $Ni(tmdt)_2$ (tmdt = trimethylenetetrathiafulvalenedithiolate), viewed along (**A**) the *b* axis and (**B**) the long molecular axis. A, B1, B2, and C denote intermolecular interactions responsible for conductivity. Reproduced from (**a**) Ref. [829] and (**b**) Ref. [831] with permission.

2.5. Dirac Electrons: Beyond Fermions

Interestingly, different fields with no obvious correlation with organic conductors have been known to catalyze the research field of organic conductors, thus broadening the scope of the field. Examples include Dirac electron systems [869–872,875,876,878,880–925] (Figure 10), which were originally a subject of particle physics but now are indicative of massless fermions in organic conductors. They occur when the materials contain a cone structure in the electronic bands, i.e., two cone-shaped bands touch at the apex of each cone, which are called Dirac points. When the Fermi levels are exactly located at the Dirac points, the materials are called zero-gap semiconductors. Because the physical properties are governed by the electrons/holes at the Fermi levels, unique behavior originating from the Dirac electrons is expected to be discovered in the zero-gap semiconductors. In fact, they were revealed in one of the oldest organic conductors, α-BEDT-TTF_2I_3, almost simultaneously by experimental and theoretical studies [883,884,886–889,891,892,895–900,902–910,916–918]. The Dirac electrons are characterized by their negligibly small masses with Fermi velocities comparable to that of light. Coupled with the research interest in topological materials [885,890,893,894,901,911–915,919], organic Dirac electron systems have become an emergent topic not only in the field of organic conductors but also in the broad field of solid-state physics. Although Dirac electron systems have also been observed in inorganic compounds, such as the famous example of graphene (awarded the Nobel Prize in Physics in 2010) [885], the advantage of crystalline organic CT salts over other kinds of materials lies in their well-defined Fermi levels based on the well-defined crystal structures, chemical compositions (stoichiometries), and the electron count of their components. Accordingly, organic Dirac systems provide a platform for intriguing particles to be produced using small glassware instead of huge accelerators. Both organic and inorganic Dirac electron systems exhibit almost temperature-independent electrical resistivity, which has been extensively studied. The reflectance spectra of such systems have also been relatively well studied [893,894,901,911,921]. In the meantime, other physical properties common to Dirac electrons, such as magnetic behavior, appear to be controversial and require further study [872,878,888,900,921]. One of the difficulties in studying organic Dirac electron systems originates from the fact that the occurrence of Dirac cones (zero-gap semiconductors) requires high pressures in most of the organic compounds, which restricts the available experiments. In this sense, recent findings regarding organic Dirac

electron systems existing at ambient pressure are important [878,920–925]. Additionally, the extremely small energy scales of organic and inorganic Dirac electron systems cause further difficulty in their study, as this makes the characteristic electronic states of zero-gap semiconductors qualitatively unclear. An understanding of the ground states of zero-gap semiconductors requires precise calculations that consider every possible interaction within the solid states as well as physical property data at the lowest temperature possible, both of which are currently being explored.

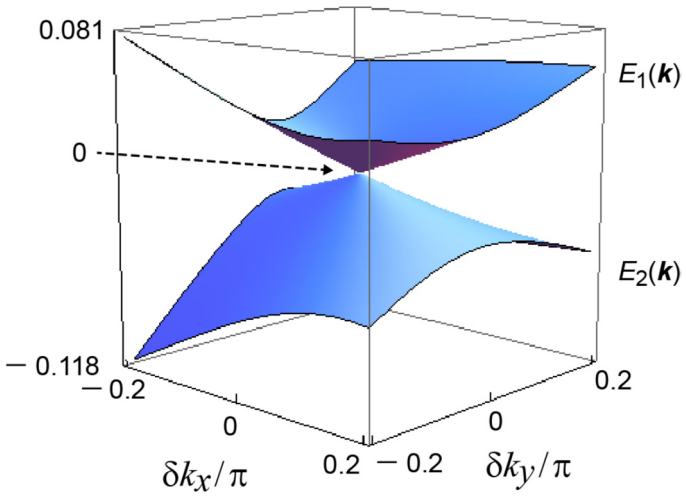


Figure 10. A characteristic curvature of the band structure, which is referred to as a Dirac cone. A close-up view around one of the two Dirac points, shown by 0 in the figure. The band structure was calculated for α-STF_2I_3. Reproduced from Ref. [920] with permission.

2.6. Chiral Conductors: Electrons in an Asymmetric Wonderland

Chirality is one of the most widely known structural features in chemistry and physics [926–958]. The control of the chirality of molecular structures and the resultant effects on their physical properties have been extensively studied for a long time. However, it has been difficult to reveal some unique conducting properties directly associated with chirality, even for single crystals belonging to non-centrosymmetric space groups [951]. Recently, an increasing number of papers on such attempts can be found [926–947,949, 950,953–959]. Chirality control is an advanced stage of "crystal designing/engineering." During the initial stages of "crystal designing" (investigated since ~1990), researchers considered and controlled the arrangement of neighboring molecules based on interatomic interactions, such as hydrogen bonding, and coordination/supramolecular chemistry [952]. Such crystal designing is based largely on molecular designing. Upon investigating the chirality control of crystal structures (investigated since ~2000), one should consider the orientation and arrangement of all the molecules in the crystals, even though there is no direct interaction between distant molecules. Thus, the synthesis of single crystals with non-centrosymmetric space groups is difficult (Figure 11), making the discovery of their unique conducting properties associated with the chirality even more so. Still, in the last two decades, there have been frequent reports on various new chiral donor molecules and their CT salts that exhibit interesting resistivity behavior with chiral crystal structures [953–959]. This demonstrates that chiral molecular (super)conductors are ever-advancing toward a new direction of organic-conductors development.

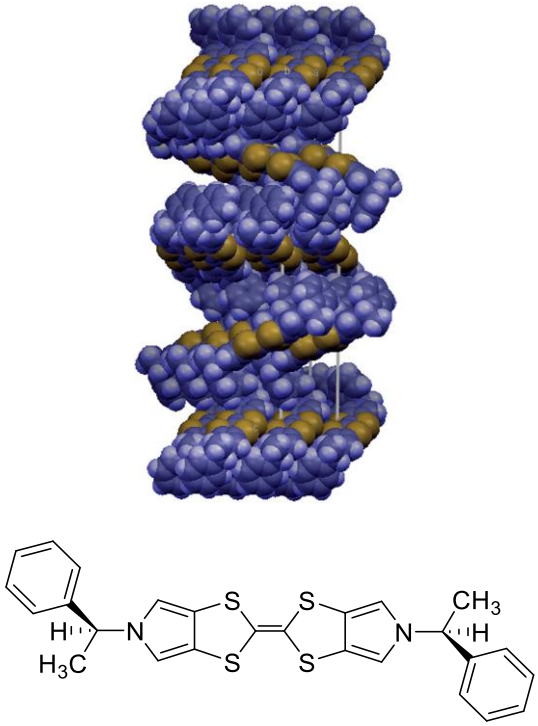


Figure 11. Organic crystal with a chiral molecular arrangement (upper; Space Group *P*1) containing a bis(pyrrolo)tetrathiafulvalene derivative (lower). The donor packing motif represents a 4_3 axis along the *c* axis (vertical). Reproduced and modified from Ref. [934] with permission from The Royal Society of Chemistry.

3. Concluding Remarks: Towards the New Age

In this review, we could refer to only a limited portion of the field of organic conductors. There are still a number of important topics that were not discussed herein. However, even from this brief review of the history of organic conductors, we can see that the field covers a wide spectrum of topics and possesses an ever-increasing potential to develop into a new field. The focus of such a field is unknown, but by maintaining close interactions with other research fields, the field of organic conductors will continue to grow and evolve, very likely until we can no longer define what "research on organic conductors" entails. Pursuing (super)conducting and magnetic properties similar to those in inorganic compounds, the early stage of organic conductors research can be characterized by the pursuit of what organic compounds can also do. In the meantime, the last two decades of the field are characterized by a new trend, i.e., the pursuit of what only organic compounds can do. Similar to some patterns of chirality, there are phenomena unique to molecules, which might produce unknown effects on conducting properties exclusively possible in molecular crystals. The new age should be a stage where the unique significance of molecular conductors is demonstrated. Without a doubt, all of the authors in this Special Issue will overjoy if this issue can help the understanding of the interesting and profound world of organic conductors, which is always ready to welcome new young generation to join us and add their new idea from different points of view.

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