



Editorial Organic Conductors

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Since the pioneering work concerning organic semiconductors in the middle of the 1900s, organic conductor research has experienced a series of milestones, from metallic to superconducting charge-transfer complexes (1980s–1990s). The history of these materials is reviewed in this Special Issue by considering a body of references [1].

The publication "Organic Conductors" covers various solids containing both organic and inorganic species with a variety of shapes and dimensions. The physical properties of these materials include electrical, magnetic, structural, optical, dielectric, and mechanical. The most prominent feature of organic conductors and related materials is the wide variety of degrees of freedom, which enables peculiar electronic states, physical properties, and phase transitions that are otherwise unobserved. As a result, even an insulating organic crystalline material can be a center of interest in this field if it provides an important piece of information concerning the mechanism of superconductivity, for example, or sheds light on other subjects of broad interest from the abovementioned point of view.

In 2022, researchers in this ever-expanding and developing field are looking for further interesting and exciting targets that are yet to be explored. On such a memorial occasion, this Special Issue, entitled "Organic Conductors", has collated 25 papers (plus editorials) from 11 countries. Readers will enjoy the latest developments in new materials, ideas, and methodologies, which will propel the field in a new direction and age.

The Special Issue consists of the following papers and reviews:

- 1. The synthesis of new molecules and organic conductors:
 - Kubo et al. reported a series of nickel–dithiolene complexes fused with bulky cycloalkane substituents to study their steric-based effects on molecular arrangements [2].
 - Kadoya et al. reported the structural and physical properties of a new organic Mott insulator with a *θ*-type molecular arrangement [3].
 - Mroweh et al. reported new chiral conducting salts based on ethylenedithiotetrathiafulvalene (EDT-TTF) derivatives, presenting their crystal structures, extended Hückel band structures, and electrical properties [4].
 - Sakaguchi et al. reported a new single-component molecular conductor, [Au(etdt)₂]·THF (etdt = ethylenedithiotetrathiafulvalenedithiolate), that is, the synthesis, structure, and physical properties of a neutral gold-dithiolenecomplex [5].
 - Akutsu et al. reported three types of new organic conductors containing HOC₂H₄SO₃⁻ anions and discussed their electrical properties in terms of the Madelung energies [6].
 - Sato et al. reported new stable neutral radical species, M^{III}(Pc)Cl₂·THF (M = Co or Fe, Pc = phthalocyanine, THF = tetrahydrofuran), as three-dimensional, single-component molecular conductors [7].
 - Koyama et al. reported the synthesis and structural, spectroscopic, and electrical properties of a new molecular conductor with a weak hydrogen-bonding network [8].



Citation: Naito, T. Organic Conductors. *Crystals* **2022**, *12*, 523. https://doi.org/10.3390/ cryst12040523

Received: 14 March 2022 Accepted: 8 April 2022 Published: 9 April 2022

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- 2. Structural, optical, magnetic, electrical, and other related properties:
 - Rohwer et al. reported the effects of deuteration on the transport properties of quasi-one-dimensional Fabre salts, (TMTTF)₂X (TMTTF = tetramethyltetrathia-fulvalene, X = Br, PF₆, and ClO₄), and discovered various conduction properties such as Mott insulators, variable range hopping, and activated band transport with a temperature-dependent bandgap [9].
 - Kitou et al. reported an experimental method for use in estimating valence electron densities, that is, frontier orbitals, in the solid state based on X-ray diffraction data [10].
 - Yoshino et al. reported the crystal structures and electrical resistivities of a series of TMTSF (TMTSF = tetramethyltetraselenafulvalene) salts with unusual stoichiometries and determined the valence state of TMTSF in each type of salt using quantum chemistry calculations [11].
- 3. Physical property measurements with new techniques and/or under extreme conditions:
 - Yamamoto et al. reported a new system for measuring the pyroelectricity of small ferroelectric single crystals [12].
 - Hino et al. reported the current- and voltage-dependence of the heat capacity of a single crystal of a charge glass compound, θ -(BEDT-TTF)₂CsZn(SCN)₄ (BEDT-TTF = bis(ethylenedithio)-tetrathiafulvalene) [13].
- 4. Spectroscopic studies concerning molecular functional crystals:
 - Hiraki et al. reported ⁷⁷Se-NMR studies on λ -type BETS superconductors, λ -(BETS)₂Fe_{1-x}Ga_xCl₄ (BETS = bis(ethyleneditho)tetraselenafulvalene), to examine the π -spin polarization affected by the localized 3*d* spins on Fe atoms [14].
- 5. Theoretical studies on organic conductors:
 - Tsumuraya et al. reported first-principle density functional theory calculations of the charge-ordered phase of α-(BEDT-TTF)₂I₃, which is closely related to Dirac electron systems [15].
 - Roy et al. reported accurate zero-temperature density matrix renormalization group calculations for κ-(BEDT-TTF)₂X, the most studied family of organic superconductors, and concluded that magnetic fluctuations in the effective half-filled band approach do not drive superconductivity in these and related materials [16].
 - Ménard et al. reported a one-dimensional alternating extended Hubbard model at quarter-filling based on a renormalization group method to examine structural instabilities in Fabre and Bechgaard salts and related organic conductors [17].
 - Suzumura et al. calculated electrical transport in nodal line semimetals of singlecomponent molecular conductors to examine the effects of acoustic phonon scattering on electrical conductivity [18].
 - Naito et al. reported a method of calculating intermolecular interactions in disordered molecular charge-transfer complexes of STF (STF = bis(ethylenedithio)diselenadithiafulvalene) by proposing a new interpretation or usage of wavefunctions [19].
 - Kesharpu et al. calculated the evolution of the shape and volume fraction of superconducting domains in relation to temperature and anion disorder in a highly anisotropic organic superconductor, (TMTSF)₂ClO₄ [20].
- 6. Molecular π -d, Dirac, and strongly correlated electron systems:
 - Cui et al. reported the high-pressure crystal structure and magnetoresistance of a single-component molecular conductor [Pt(dddt)₂] (dddt = 5,6-dihydro-1,4-dithiin-2,3-dithiolate), a theoretically expected three-dimensional Dirac electron system, under high pressure [21].
 - Yasuzuka reviewed the interplay between the vortex dynamics and superconducting gap structures in layered organic superconductors containing strongly correlated electron systems. Particular attention was paid to the symmetry of

the superconducting gaps, which is a key feature in understanding the pairing mechanism [22].

- 7. Organic magnets, dielectrics, semiconductors, superconductors, and photoconductors:
 - Manabe et al. reported reversible structural and magnetic transformations in inorganic ladder compounds; these transformations are dependent on the gas-phase chemical species absorbed in single crystals [23].
- 8. Molecular electronics, optoelectronics, spintronics, devices, and related topics:
 - Kawasugi et al. reported doping effects using field-effect transistors consisting of electric double layers of charge-ordered insulators, α-(BEDT-TTF)₂I₃, and α-(BETS)₂I₃ [24].
 - Gou et al. reported experimental and modelling studies of a series of highperformance inorganic semiconductor devices based on doped GaAs and related materials [25].

The Guest Editor would like to express his sincere thanks to all of the authors for their contributions to this Special Issue, as well as to all of the reviewers for the time and effort expended to provide valuable feedback to the authors. Special thanks also go to the *Crystals* editorial office members, Ms. Debbie Yang in particular, for their continuous collaboration, timely communication, and efficient support, along with their friendly and professional attitude; without their contributions, the publication of this Special Issue would not have been possible. The Guest Editor hopes that this Special Issue will encourage more and more scientists to join this field to further expand the horizon and to discover new possibilities of molecular conductors.

Funding: The author is grateful for the financial support from a Grant-in-Aid for Challenging Exploratory Research (18K19061) of JSPS, the Iketani Science and Technology Foundation (ISTF; 0331005-A), the Research Grant Program of the Futaba Foundation, the Casio Science Promotion Foundation, and an Ehime University Grant for Project for the Promotion of Industry/University Cooperation.

Conflicts of Interest: The author declares no conflict of interest.

References

- 1. Naito, T. Modern History of Organic Conductors: An Overview. Crystals 2021, 11, 838. [CrossRef]
- Kubo, K.; Sadahiro, M.; Arata, S.; Hoshino, N.; Kadoya, T.; Akutagawa, T.; Kato, R.; Yamada, J.-I. Donor-Type Nickel–Dithiolene Complexes Fused with Bulky Cycloalkane Substituents and Their Application in Molecular Conductors. *Crystals* 2021, 11, 1154. [CrossRef]
- Kadoya, T.; Sugiura, S.; Higashino, T.; Tahara, K.; Kubo, K.; Sasaki, T.; Takimiya, K.; Yamada, J.-I. Dihedral-Angle Dependence of Intermolecular Transfer Integrals in BEDT-BDT-Based Radical-Cation Salts with *θ*-Type Molecular Arrangements. *Crystals* 2021, 11, 868. [CrossRef]
- 4. Mroweh, N.; Auban-Senzier, P.; Vanthuyne, N.; Lopes, E.B.; Almeida, M.; Canadell, E.; Avarvari, N. Chiral Conducting Me-EDT-TTF and Et-EDT-TTF-Based Radical Cation Salts with the Perchlorate Anion. *Crystals* **2020**, *10*, 1069. [CrossRef]
- Sakaguchi, K.; Zhou, B.; Idobata, Y.; Kamebuchi, H.; Kobayashi, A. Syntheses, Structures, and Physical Properties of Neutral Gold Dithiolate Complex, [Au(etdt)₂]·THF. Crystals 2020, 10, 1001. [CrossRef]
- Akutsu, H.; Koyama, Y.; Turner, S.S.; Furuta, K.; Nakazawa, Y. Structures and Properties of New Organic Conductors: BEDT-TTF, BEST and BETS Salts of the HOC₂H₄SO₃⁻ Anion. *Crystals* 2020, *10*, 775. [CrossRef]
- Sato, R.; Matsuda, M. Formation of Three-Dimensional Electronic Networks Using Axially Ligated Metal Phthalocyanines as Stable Neutral Radicals. Crystals 2020, 10, 747. [CrossRef]
- Koyama, S.; Kawai, M.; Takaishi, S.; Yamashita, M.; Hoshino, N.; Akutagawa, T.; Kanno, M.; Iguchi, H. Synthesis, Structure and Physical Properties of (trans-TTF-py₂)_{1.5}(PF₆)·EtOH: A Molecular Conductor with Weak CH…N Hydrogen Bondings. *Crystals* 2020, 10, 1081. [CrossRef]
- Rohwer, A.; Dressel, M.; Nakamura, T. Deuteration Effects on the Transport Properties of (TMTTF)₂X Salts. *Crystals* 2020, 10, 1085. [CrossRef]
- 10. Kitou, S.; Hosogi, Y.; Kitaura, R.; Naito, T.; Nakamura, T.; Sawa, H. Direct Observation of Molecular Orbitals Using Synchrotron X-ray Diffraction. *Crystals* **2020**, *10*, 998. [CrossRef]
- 11. Yoshino, H.; Iwasaki, Y.; Tanaka, R.; Tsujimoto, Y.; Matsuoka, C. Crystal Structures and Electrical Resistivity of Three Exotic TMTSF Salts with I 3–: Determination of Valence by DFT and MP2 Calculations. *Crystals* **2020**, *10*, 1119. [CrossRef]

- Yamamoto, K.; Kawasaki, A.; Chinen, T.; Ryugo, K. Temperature-Modulated Pyroelectricity Measurements of a Thin Ferroelectric Crystal with In-Plane Polarization and the Thermal Analysis Based on One-Dimensional Layer Models. *Crystals* 2021, *11*, 880. [CrossRef]
- Hino, K.; Nomoto, T.; Yamashita, S.; Nakazawa, Y. Single Crystal Heat Capacity Measurement of Charge Glass Compound θ-(BEDT-TTF)₂CsZn(SCN)₄ Performed under Current and Voltage Application. *Crystals* 2020, 10, 1060. [CrossRef]
- Hiraki, K.; Takahashi, T.; Akiba, H.; Nishio, Y.; Zhou, B. Microscopic Observation of π Spin Polarization by *d* Localized Spin in λ Type BETS Based Organic Superconductors. *Crystals* 2020, 10, 1055. [CrossRef]
- 15. Tsumuraya, T.; Seo, H.; Miyazaki, T. First-Principles Study on the Stability and Electronic Structure of the Charge-Ordered Phase in *α*-(BEDT-TTF)₂I₃. *Crystals* **2021**, *11*, 1109. [CrossRef]
- Roy, D.; Clay, R.T.; Mazumdar, S. Absence of Superconductivity in the Hubbard Dimer Model for κ-(BEDT-TTF)₂X. *Crystals* 2021, 11, 580. [CrossRef]
- 17. Ménard, M.; Bourbonnais, C. One-Dimensional Alternating Extended Hubbard Model at Quarter-Filling and Its Applications to Structural Instabilities of Organic Conductors. *Crystals* **2020**, *10*, 942. [CrossRef]
- Suzumura, Y.; Kato, R.; Ogata, M. Electric Transport of Nodal Line Semimetals in Single-Component Molecular Conductors. Crystals 2020, 10, 862. [CrossRef]
- 19. Naito, T.; Suzumura, Y. Theoretical Model for Novel Electronic State in a Dirac Electron System Close to Merging: An Imaginary Element between Sulphur and Selenium. *Crystals* **2022**, *12*, 346. [CrossRef]
- 20. Kesharpu, K.K.; Kochev, V.D.; Grigoriev, P.D. Evolution of Shape and Volume Fraction of Superconducting Domains with Temperature and Anion Disorder in (TMTSF)₂ClO₄. *Crystals* **2021**, *11*, 72. [CrossRef]
- Cui, H.; Yeung, H.H.-M.; Kawasugi, Y.; Minamidate, T.; Saunders, L.K.; Kato, R. High-Pressure Crystal Structure and Unusual Magnetoresistance of a Single-Component Molecular Conductor [Pd(dddt)₂] (dddt = 5,6-dihydro-1,4-dithiin-2,3-dithiolate). *Crystals* 2021, 11, 534. [CrossRef]
- 22. Yasuzuka, S. Interplay between Vortex Dynamics and Superconducting Gap Structure in Layered Organic Superconductors. *Crystals* **2021**, *11*, 600. [CrossRef]
- Manabe, J.; Nishida, K.; Zhang, X.; Nakano, Y.; Fujibayashi, M.; Cosquer, G.; Inoue, K.; Shimono, S.; Ishibashi, H.; Kubota, Y.; et al. Gas-Dependent Reversible Structural and Magnetic Transformation between Two Ladder Compounds. *Crystals* 2020, 10, 841. [CrossRef]
- Kawasugi, Y.; Masuda, H.; Pu, J.; Takenobu, T.; Yamamoto, H.M.; Kato, R.; Tajima, N. Electric Double Layer Doping of Charge-Ordered Insulators α-(BEDT-TTF)₂I₃ and α-(BETS)₂I₃. *Crystals* 2021, 11, 791. [CrossRef]
- Gou, Y.; Wang, J.; Cheng, Y.; Guo, Y.; Xiao, X.; Liu, H.; Tan, S.; Zhou, L.; Yang, H.; Deng, G.; et al. Experimental and Modeling Study on the High-Performance p⁺⁺-GaAs/n⁺⁺-GaAs Tunnel Junctions with Silicon and Tellurium Co-Doped InGaAs Quantum Well Inserted. *Crystals* 2020, 10, 1092. [CrossRef]