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Editorial

Functional Molecular Materials Insights

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In the commemorative Special Issue titled "Insights into Functional Molecular Materials—A Themed Collection Honoring Professor Manuel Almeida on His 70th Birthday", eminent researchers from around the globe in the field of molecular materials science come together to acknowledge and celebrate the notable scientific contributions of Professor Almeida.

The topics covered in the published articles exemplify the interdisciplinary nature of this scientific domain, seamlessly integrating preparative chemistry and condensed matter physics in equal proportions. This unique Special Issue features fifteen insightful contributions, and a brief overview of these contributions is presented below.

The first paper, "Synthesis and Structural and Magnetic Properties of Polycrystalline $GaMo_4Se_8$" by J.F. Malta and A.P. Gonçalves from IST, the University of Lisboa (Portugal), and M.S.C. Henriques and J.A. Paixão from the University of Coimbra (Portugal), reports an innovative two-step synthesis of pure polycrystalline $GaMo_4Se_8$, a lacunar spinel belonging to the GaM_4X_8 family. Phase purity and composition are confirmed through XRD and SEM analyses. Magnetic investigations reveal the presence of cycloidal, skyrmionic, and ferromagnetic phases, contributing valuable insights into the compound's magnetic behavior and potential applications (contribution 1).

The second paper by H. Tajima and T. Kadoya from the Japanese Universities of Hyogo and Konan investigates "Nonthermal Equilibrium (NTE) Process of Charge Carrier Extraction in Metal/Insulator/Organic Semiconductor/Metal (MIOM) Junction" with Schottky-type contacts, contrasting it with standard capacitors. Strategies like ohmic contacts or high-mobility organic semiconductors are suggested to mitigate NTE's negative impact in organic field-effect transistors. The NTE process could find applications in OFETs as memory devices and for finding charge injection barriers (contribution 2).

The contribution by R. Kato from the RIKEN Laboratory at Wako, Saitama, and T. Tsumuraya from Kumamoto University in Japan explores "Dirac Cone Formation in Single-Component Molecular Conductors Based on Metal Dithiolene Complexes", using tight-biding models and first-principles density functional theory (DFT) calculations. The tight-binding model predicts the emergence of Dirac cones in the studied systems, which is associated with a stretcher bond type of molecular arrangement (contribution 3).

The paper "On the Size of Superconducting Islands on the Density-Wave Background in Organic Metals" by V.D. Kochev, S.S. Seidov, and P.D. Grigoriev from the National University of Science and Technology "MISiS" (Russia) and the L.D. Landau Institute for Theoretical Physics (Russia) explores spatial inhomogeneity in high-temperature organic superconductors, specifically the transition from superconductivity to a density wave state in quasi-one-dimensional metals with imperfect nesting. External pressure influences this transition by changing electron dispersion. By estimating the size of superconducting islands during this transition, the authors provide insights into spatial heterogeneity in organic superconductors (contribution 4).

In a paper entitled "Band Structure Evolution during Reversible Interconversion between Dirac and Standard Fermions in Organic Charge-Transfer Salts" by R. Oka, K.

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Ohara, K. Konishi, and T. Naito from Ehime University (Japan) and I. Yamane and T. Shimada from Hokkaido University (Japan), organic charge-transfer salts studied, α-D_2I_3 (D = ET, BETS), exhibit temperature-sensitive charge-transfer interactions, transforming flat-bottomed bands into Dirac cones with decreasing temperature. The observed band reshaping under ambient pressure provides insights into the lifecycle of Dirac fermions (DFs). While shedding light on DF systems, the study emphasizes the need for further research to deepen our understanding of the nature of DFs (contribution 5).

"Driving a Molecular Spin-Peierls System into a Short Range Ordered State through Chemical Substitution" explores the effect of the introduction of bromine atoms on quasi-1D spin-Peierls system potassium TCNQ (TCNQ = 7,7,8,8-tetracyanoqunidimethane). The resulting derivative, potassium $TCNQBr_2$, shows evidence of residual magnetism, suggesting short-range and potentially disordered correlations. While magnetic susceptibility data hint at 1D behavior, muon spin spectroscopy reveals a departure from the expected spin-Peierls system behavior, indicating a dominance of short-range magnetic correlations. This study by A. Berlie from the Rutherford Appleton Laboratory, at Harwell Campus in Oxfordshire (UK) and I. Terry and M. Szablewski from Durham University (UK) suggests that the bulky bromine atoms prevent the structural changes required for the system to undergo a spin-Peierls transition (contribution 6).

In paper number seven entitled "Two One-Dimensional Copper-Oxalate Frameworks with the Jahn–Teller Effect: $[(CH_3)_3NH]_2[Cu(\mu\text{-}C_2O_4)(C_2O_4)]\cdot 2.5H_2O$ (I) and $[(C_2H_5)_3NH]_2[Cu(\mu\text{-}C_2O_4)(C_2O_4)]\cdot H_2O$ (II)", B. Zhang, Y. Sun, T. Liang, M. Liu, and D. Zhu from the Chinese Academy of Sciences and Y. Zhang and Z. Wang from Peking University, China, reported that these salts feature Jahn–Teller-distorted copper ions in an octahedral coordination and that the crystal structure is characterized by hydrogen bonds among ammonium, water, and the copper–oxalate framework, creating a 3D network. Both salts are insulators and exhibit ferromagnetic and weak-ferromagnetic behaviors with no long-range ordering observed above 2 K (contribution 7).

The contribution "A Simulation Independent Analysis of Single- and Multi-Component cw ESR Spectra" by A. S. Roy, B. Dzikovski, and M. Srivastava from Cornell University (USA), D. Dolui and A. Dutta from the Indian Institute of Technology Bombay (India), and O. Makhlynets from Syracuse University (USA) introduces a novel simulation-independent approach for the precise analysis of continuous-wave electron spin resonance (cw ESR) spectra, crucial for understanding free radicals and paramagnetic metal complexes. The method, based on wavelet packet transform, accurately extracts spectral information, overcoming challenges posed by poorly resolved spectra. Applied to various systems, this approach demonstrates consistency and accuracy, even in identifying the features of a 5% minor component in a two-component system. The method's efficacy was validated with well-studied systems (contribution 8).

A paper by Q. Wan, M. Wakizaka, H. Zhang, N. Funakoshi, S. Takaishi, and M. Yamashita from Tohoku University (Japan), Y. Shen from Xi'an Jiaotong University (China), and C.-M. Che from The University of Hong Kong (China), entitled "*A New Organic Conductor of Tetramethyltetraselenafulvalene (TMTSF) with a Magnetic Dy(III) Complex*", reports the synthesis of $(TMTSF)_5[Dy(NCS)_4(NO_3)_2]CHCl_3$ using the electrochemical oxidation method. This salt shows a semiconducting behavior with a conductivity of 0.2 $S\cdot cm^{-1}$ at room temperature and an activation energy of 34 meV at ambient pressure. This preliminary study provides information for designing new hybrid materials based on molecular conductors and polyvalent magnetic 4f metal complexes (contribution 9).

The study "Superconductivity and Fermi Surface Studies of β″-$(BEDT\text{-}TTF)_2[(H_2O)(NH_4)_2Cr(C_2O_4)_3]\cdot$18-Crown-6" presents radiofrequency penetration depth measurements on a 2D organic superconductor with the largest layer separation between consecutive conduction layers, using a contactless tunnel diode oscillator measurement technique. Measurements reveal its behavior under different orientations to the crystal conduction planes. When parallel to the layers, $Hc2$ is 7.6 T with no signs of inhomogeneous superconductivity. Perpendicular orientation shows Shubnikov–de Haas oscillations,

indicating high anisotropy in $Hc2$, particularly a low $Hc2\perp$ of 0.4 T, possibly due to a lower effective mass. The work was developed by B. Laramee, R. Ghimire, and C.C. Agosta from Clark University (Worcester, MA, USA), D. Graf from the National High Magnetic Field Laboratory (Florida State University, USA), and L. Martin and T.J. Blundell from Nottingham Trent University (UK) (contribution 10).

The contribution "Vibronic Relaxation Pathways in Molecular Spin Qubit $Na_9[Ho(W_5O_{18})_2]\cdot 35H_2O$ under Pressure", authored by J.L. Musfeldt from the University of Tennessee (USA), Z. Liu from the University of Illinois Chicago (USA), and D. López-Alcalá, Y. Duan, A. Gaita-Ariño, J.J. Baldoví, and E. Coronado from the University of Valencia (Spain), investigates controlling spectral sparsity and decoherence in a qubit system using diamond anvil cell techniques, infrared spectroscopy, and first-principles calculations. The results suggest that applying negative pressure through chemical means or strain could improve transparency in the spin qubit system, offering potential for better managing decoherence in quantum devices (contribution 11).

The paper "Giant Angular Nernst Effect in the Organic Metal α-(BEDT-TTF)$_2$KHg(SCN)$_4$" by D. Krstovska from Ss. Cyril and Methodius University (North Macedonia), E.S. Choi from Florida State University (USA), and E. Steven from Jakarta Utara DKI (Indonesia) reports a substantial Nernst effect in the charge density wave state of this organic metal. Momentum relaxation dynamics in the low-field CDW state indicate significant carrier mobility, contributing to the large Nernst signal. However, this effect diminishes in the high-field CDW state, where only phonon drags and electron–phonon interactions contribute to the thermoelectric signal. These findings challenge previous understandings of the complex properties of this organic metal (contribution 12).

The metal–insulator transition in κ-(BEDT-TTF)$_2$Hg(SCN)$_2$Br at $T_{MI} \approx 90$ K involves a crystal shift from monoclinic to triclinic. The triclinic phase tends towards a Mott insulating state, causing increased resistance below T_{MI}, which is suppressed by external pressure. This remarkable "Effect of External Pressure on the Metal–Insulator Transition of the Organic Quasi-Two-Dimensional Metal κ-(BEDT-TTF)$_2$Hg(SCN)$_2$Br" is reported in this paper by S.I. Pesotskii, R.B. Lyubovskii, G. V. Shilov S.A. Torunova, V.N. Zverev, and E.I. Zhilyaeva from the Russian Academy of Sciences (Russia) and E. Canadell from ICMAB-CSIC (Spain). Quantum oscillations align with the calculated Fermi surface for the triclinic phase, explaining the material's behavior around 100 K. Notably, differences are observed in the behaviors of the isostructural salts κ-(BEDT-TTF)$_2$Hg(SCN)$_2$Br and κ-(BEDT-TTF)$_2$Hg(SCN)$_2$Cl (contribution 13).

"Spin-Peierls, Spin-Ladder and Kondo Coupling in Weakly Localized Quasi-1D Molecular Systems: An Overview", by J.-P. Pouget from Université Paris-Saclay (France), explores magneto-structural properties in quasi-one-dimensional molecular organics with electron–electron correlations, emphasizing spin–charge decoupling and singlet dimer formation. Examples like (TMTTF)$_2$X Fabre salts and Per$_2$-M(mnt)$_2$ systems illustrate the spin-Peierls instabilities and the 3D-SP ground states. This study also delves into the unique features of correlated 1D systems, including coexisting orders and soliton nucleation in perturbed spin-Peierls systems (contribution 14).

"Lanthanide-Based Metal–Organic Frameworks with Single-Molecule Magnet Properties" allow for tunable magnetic behaviors through factors like solvent, temperature, and organic linkers. This overview covers synthetic methods and strategies, including redox activity and chirality, for controlling SMM behavior in Ln-MOFs. The discussion also touches on intriguing phenomena like the CISS effect and CPL. The paper was authored by F. Manna, M. Oggianu, and M.L. Mercuri from the University of Cagliari (Italy) and N. Avarvari from the University of Angers (France) (contribution 15).

This issue will provide valuable insights into the rapidly evolving landscape of current research in molecular materials and related studies. We would like to sincerely thank all the authors who contributed to this Special Issue for their dedicated efforts and the outstanding quality of their submissions. Finally, we would like to express our gratitude

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