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Review

Metallosupramolecular approach toward multifunctional magnetic devices for molecular spintronics☆

María Castellano[a,1], Rafael Ruiz-García[a,b], Joan Cano[a,b], Jesús Ferrando-Soria[a,2], Emilio Pardo[a], Francisco R. Fortea-Pérez[a], Salah-Eddine Stiriba[a,c], Wdeson P. Barros[d], Humberto O. Stumpf[d], Laura Cañadillas-Delgado[e], Jorge Pasán[f], Catalina Ruiz-Pérez[f], Giovanni de Munno[g], Donatella Armentano[g], Yves Journaux[h,i], Francesc Lloret[a,*], Miguel Julve[a,*]

[a] *Instituto de Ciencia Molecular (ICMol), Universitat de València, Paterna, València, Spain*
[b] *Fundació General de la Universitat de València (FGUV), València, Spain*
[c] *Equipe de Chimie Moléculaire, Matériaux et Modélisation-C3M, Faculté Polydisciplinaire de Safi, Université Cadi Ayyad, Safi, Morocco*
[d] *Laboratório de Química de Materiais Moleculares (LQMMol), Departamento de Química (ICEx), Universidade Federal de Minas Gerais, Belo Horizonte, Brazil*
[e] *Centro Universitario de la Defensa de Zaragoza, 50090 Zaragoza, Spain*
[f] *Laboratorio de Rayos X y Materiales Moleculares (MATMOL), Departamento de Física, Facultad de Ciencias (Sección Física), Universidad de La Laguna, Tenerife, Spain*
[g] *Centro di Eccellenza CEMIF.CAL, Dipartimento di Chimica, Università della Calabria, 87030 Cosenza, Italy*
[h] *Institut Parisien de Chimie Moléculaire, UPMC Univ Paris 06, Paris, France*
[i] *CNRS, UMR 7071, Paris, France*

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Abbreviations: EE, electron exchange; ET, electron transfer; MAC, molecular antiferromagnetic coupler; MFC, molecular ferromagnetic coupler; MMC, molecular magnetic capacitor; MMR, molecular magnetic rectifier; MMS, molecular magnetic switch; MMW, molecular magnetic wire; OA, oligo(acene); OAQ, oligo(acenequinone); OP, oligo(phenylene); OPE, oligo(phenylene-ethynylene); QUBIT, quantum bit; QCA, quantum cellular automata; SCU, spin-containing unit.

☆ Supramolecular coordination chemistry of aromatic polyoxalamide ligands. Part 2.

* Corresponding authors.
E-mail addresses: francisco.lloret@uv.es (F. Lloret), miguel.julve@uv.es (M. Julve).
[1] Present address: Laboratoire de Physique de la Matière Condensée, CNRS École Polytechnique, 91128 Palaiseau, France.
[2] Present address: Department of Chemistry, University of Manchester, M13 9PL Manchester, UK.

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ABSTRACT

The work presented in this review constitutes a successful extension of our group's research on the chemistry and physics of dinuclear copper(II) metallacyclophanes with aromatic polyoxalamide ligands. The design and synthesis of metallacyclic complexes that contain multiple electro- and photoactive (either metal- or ligand-based) spin carriers and the study of their spectroscopic and magnetic properties as well as their redox and photochemical activity are of large interest in the multidisciplinary field of metallosupramolecular chemistry. In doing this, a ligand design approach has been followed which is based on the copper(II)-mediated self-assembly of bis(oxamato) bridging ligands possessing potentially electro- and photoactive, extended π-conjugated aromatic spacers. This strategy benefits from the inherent physical and chemical properties of aromatic organic molecules by functionalizing them with two oxamato donor groups to get dinucleating ligands that are then able to self-assemble with square planar Cu^{II} ions affording the targeted oxamato-based dicopper(II) metallacyclophanes. The organic functionalization in this new class of metallacyclic systems constitutes a unique example of ligand design for the supramolecular control of the structure and magnetic properties, as well as the electro- and photochemical activities. This novel class of oxamato-based dicopper(II) metallacyclophanes provides excellent models for the fundamental study on through-ligand long-distance and redox- or photo-triggered electron exchange phenomena, which are two central topics in molecular magnetism and molecular electronics. Using these simple dinuclear metallacyclic complexes as dynamic chemical systems to perform specific and selective tasks under the control of an external (electro- and/or photochemical) stimulus that switches "ON" and "OFF" their electronic (optical and/or magnetic) properties may have an enormous impact in several domains of molecular nanoscience. Hence, oxamato-based dicopper(II) metallacyclophanes appear as very promising candidates to get multifunctional magnetic devices controlling and facilitating the spin communication ("molecular magnetic couplers" and "molecular magnetic wires") or exhibiting charge storage ("molecular magnetic capacitors") and bistable spin behavior ("molecular magnetic rectifiers" and "molecular magnetic switches") for potential applications in information processing and storage in the emerging areas of molecular spintronics and quantum computing. Moreover, because of the potential high affinity for a variety of metal surfaces through the free carbonyl-oxygen atoms of the oxamate groups, they are very appealing candidates for the study of coherent electron transport through single molecules.

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1. Introduction

1.1. Toward molecular spintronics and quantum computation through molecular magnetism and molecular electronics

Molecular spintronics is a new and emergent field of spintronics that can benefit from the well-established developments in molecular magnetism and molecular electronics [1–3]. As occurs in these two related fields, molecular spintronics can be divided into two areas, namely single-molecule spintronics and molecule-based spintronics [4]. The first area focuses on the study of the spin-dependent electron transport through a single molecule and the use of these individual molecules as active components of spintronic circuits [4c,d]. The second area deals with the design and synthesis of new materials from molecular precursors ('molecule-based materials') that can mimic the unique magnetic and electronic properties exhibited by the existing conventional inorganic materials used in spintronics [4b,e].

Single-molecule spintronics is considered as one promising way for miniaturization of electronic circuits leading to highly integrated magnetic and electronic systems [5–8]. In order to build up a molecule-based spintronic circuit, it is necessary to generate and combine many molecular magnetic components, such as spin wires, switches, rectifiers, diodes, capacitors, valves, or transistors [5], whose non-magnetic analogs are so familiar to molecular electronics [6]. Quantum 'bits' (QUBITs) and quantum 'dot' cellular automata (QCA) are other two remarkable examples of molecular spintronic devices which would constitute the basic components for the construction of more complex magnetic logic systems for quantum computation [7,8].

Depending on the nature of the charge and spin carrier, either organic radicals or transition metal ions, two different organic- and metal-based approaches to single-molecule spintronics are known. The magnetic coupling through extended π-conjugated aromatic spacers was investigated in semiquinone, nitronyl nitroxide, methylene, and verdazyl diradicals, which may constitute illustrative examples of spin wires [9]. Besides, Irie and others have reported illustrative examples of spin switches that are based on nitronyl nitroxide diradicals [10]. On the other hand, Shultz and others have systematically investigated the nature of the magnetic and electronic coupling in transition metal complexes of simple semiquinone diradicals or mixed semiquinone-nitronyl nitroxide donor–acceptor diradicals, which may be considered a metal–organic approach to single-molecule spintronics [11]. However, these purely organic or hybrid metal–organic strategies are severely limited by the low stability of most of the organic radicals, a feature which reduces dramatically the number of available building blocks for molecular spintronic devices. In contrast, polymetallic complexes are rather stable entities and they thus appear as a potential alternative to molecular spintronic devices.

In the following section, a brief historical perspective is given on the magnetic and electronic communication in polymetallic (coordination and organometallic) complexes to introduce the current

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