This book aims to provide a coherent and pedagogical collection of articles on the physics and applications of molecular magnets. All contributors have played a major role in either (1) discovering or elucidating the physics that underlies molecular magnets, or in (2) the present exploration of avenues toward their applications. Issues that are by now well understood as well as open questions are covered. Inevitably, overlaps among some chapters do occur, but we are sure that the reader will find them complementary rather than repetitious.

Molecular magnets are made up of chemically identical molecules with high-spin cores. The cornerstone for the rise of present day interest in molecular magnetism was the discovery of magnetic quantum tunneling in Mn\(_{12}\)-acetate molecules. There was before 1996 some indirect evidence for quantum tunneling of large spins. In 1993, magnetic hysteresis (and thus magnetic memory) at liquid Helium temperatures was shown to come from single molecular clusters of Mn\(_{12}\)-acetate. However, a clear imprint of magnetic quantum tunneling was only observed in 1996. Then, experiments revealed that magnetic hysteresis in Mn\(_{12}\)-acetate is rather unconventional, in that the magnetization jumps at equally spaced values of the applied magnetic field. The gist of this effect is that spins can tunnel between different magnetic states as they are brought on and off resonance by an external magnetic field. Mn\(_{12}\)-acetate molecules thus behave as “single molecule magnets” (SMMs). “Resonant spin tunneling” in molecular magnets illustrates beautifully quantum physics at the mesoscopic scale, that is, in the crossover region between the macroscopic and microscopic worlds, where quantum and classical physics meet. Finally, SMMs are a variant of magnetic nanoparticles, which are at the basis of magnetic recording. Much interest in SMMs arises from this fact.

The field has expanded considerably in the last two decades, owing to the creativity of molecular chemists (who have crafted high and low spin clusters and single chain magnets), to the observation and elucidation of interesting phenomena (e.g., hole burning, spin avalanches and deflagration, as well as dipolar long-range ordering), and to the development of experimental techniques (e.g., single molecule manipulation on substrates). Finally, there is the vibrant ongoing work on applications. Most of it has to do with the fact that single molecule magnets are potential
2-level qubits for quantum computation. There are other applications for molecular magnets, such as to magnetic refrigeration (making use of the magneto-caloric effect) of electronic devices at cryogenic temperatures.

A brief historical account of the discovery of stepwise hysteresis loops, which are the hallmark of molecular magnets, as well as the physics of the underlying magnetic-quantum-tunneling phenomena, can be read in the first section, *Tunneling of Single Molecule Magnets*. How stepwise hysteresis loops were discovered, and first reported early in 1996, is the subject of Chap. 1. How the existence of stepwise hysteresis loops was subsequently corroborated in Mn$_{12}$-acetate single crystals, and more, can be read in Chap. 2. The theory of magnetic quantum tunneling that takes orbital angular momentum into account is given in Chap. 3. There is however more in the first section. Interesting effects that cannot be accounted for assuming each SMM acts as a single spin S are reported and explained in Chap. 4.

The second section, *Beyond Single Molecules*, covers various collective phenomena. Deflagration is one of them. It has been found to proceed in molecular magnets by rapidly moving magnetic-quantum-tunneling fronts, much as ordinary deflagration takes place by chemical combustion processes. Experimental and theoretical accounts are given in Chaps. 5 and 6, respectively. A rather different sort of collective phenomenon, equilibrium magnetic phase transitions, have been observed in some of the best known molecular magnets. Magnetic ordering is brought about by magnetic-dipolar interactions. Because system-wide ordering processes cannot bypass slow quantum tunneling processes, the realization of magnetic ordering was not a foregone conclusion. Order can either be destroyed by heating, through a classical phase transition, or by applying a transverse magnetic field, through a quantum phase transition. This is the subject of Chap. 7. *Single Chain Magnets*, the subject of Chap. 8, resemble SMMs in that they can relax extremely slowly. Their underlying physics is however rather different. In single chain magnets relaxation proceeds through thermal excitation of domain walls. Models are also discussed in this chapter. Metal-phthalocyanine (MPc) are uniquely suited for the exploration of the intrinsic mechanisms which give rise to molecular magnetism. The structural and magnetic properties of bulk crystals, thin films and single MPcs molecules adsorbed on different substrates are covered in Chap. 9. The Kondo interaction, tunneling processes, switchability and spin control are reviewed.

Most of the section on *Applications* is devoted to issues that arise from the role molecular magnets can play in information technology. How to control and exploit the quantum properties of SMMs, achievements of recent years and foresight for their near future are all weaved into *Molecular Nanomagnets for Information Technologies*, which is Chap. 10. In Chap. 11, *Molecular Magnets for Quantum Information Processing*, a brief introduction into quantum computing is given. Di-Vincenzo’s criteria for its successful physical implementation are introduced and used as a guideline throughout. Utilization and control (mainly, through the spin-electric effect) of the spin degrees of freedom in SMMs as qubit states is considered. The various decoherence mechanisms which affect SMMs and their advantages on this point over more traditional qubits are examined. Finally, a proposal to implement Grover’s algorithm using molecular magnets is discussed. In Chap. 12,
Single Molecule Spintronics, recently developed techniques that can be applied to measurements of electronic transport through a SMM are discussed. Spectroscopic information, obtained from measurements on spin-transistor-like three-terminal set ups, confirms the high-spin state and magnetic anisotropy of the robust Fe$_4$ SMM. The experimental observation that electric gate fields drastically modify the magnetic properties of an oxidized or reduced molecule is discussed. The main aim of molecular quantum spintronics, that is, to bring together concepts from spintronics, molecular electronics and quantum computing for the purpose of fabrication, characterization, and study of molecular devices—such as, molecular spin-transistors and molecular spin-valves—are reviewed in Chap. 13 (Molecular Quantum Spintronics). Finally, Chap. 14 is devoted to a totally different topic, the application (by means of the magnetocaloric effect) of molecular magnets to very low temperature refrigerants in microdevices.

In closing, the Editors wish to express their pleasure at having worked with the authors, and we would like to thank each and everyone of them for their warm response and full co-operation.

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