

Photogenerated Radical Qubits and Their Spin Polarization Properties in Acene-Based Metal-Organic Frameworks

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<https://hdl.handle.net/2324/7182434>

出版情報：九州大学, 2023, 博士（工学）, 課程博士
バージョン：
権利関係：やむを得ない事由により本文ファイル非公開（4）



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論 文 名 : Photogenerated Radical Qubits and Their Spin Polarization Properties
in Acene-Based Metal-Organic Frameworks
(アセンベースの有機金属骨格における光生成ラジカル量子ビットと
そのスピン分極特性)

区 分 : 甲

論 文 内 容 の 要 旨

Generating and controlling electron spin polarization (ESP) is crucial in the foundation of a wide range of spin-related technologies. In the context of quantum information science (QIS), it provides the spin systems necessary for qubit operation and it also leads to highly sensitive quantum sensing. Dynamic nuclear polarization (DNP), which transfers the polarization from electron spins to nuclear spins, can lead to increased sensitivity in nuclear magnetic resonance and magnetic resonance imaging. ESP using molecular materials has attracted particular attention because of their high polarization performance and atomic-scale controllability. In addition to ESP, long spin-lattice relaxation times (T_1) are important for spin manipulations and highly efficient DNP. If ESP with a long T_1 at room temperature can be achieved, quantum sensing and DNP at the temperature where biomolecules are actually active would be realized. The development of molecular materials exhibiting ESP with a longer T_1 at room temperature will be the key to expanding the scope of future molecular quantum technology. In addition, the extended coherent time (T_2) is necessary to improve the application for quantum sensing. In this work, we report the realization of photogenerated spin-polarized radicals with extremely long T_1 and T_2 at room temperature in a MOF in which azaacene chromophores are densely integrated.

In Chapter 2, the generation of spin polarization is key in quantum information science and dynamical nuclear polarization. Polarized electron spins with long spin-lattice relaxation times (T_1) at room temperature are important for these applications but have been difficult to achieve.

We report the realization of spin-polarized radicals with extremely long T_1 at room temperature in a metal-organic framework (MOF) in which azaacene chromophores are densely integrated. Persistent radicals are generated in the MOF by charge separation after photoexcitation. Spin polarization of triplet generated by photoexcitation are successfully transferred to the persistent radicals. Pulse ESR measurements reveal that the T_1 of the polarized radical in the MOF is as long as 214 μs and relatively long spin-spin relaxation time T_2 of the radicals up to 0.98 μs at room temperature. The achievement of extremely long spin polarization in MOFs with nanopores accessible to guest molecules will be an important cornerstone for future highly sensitive quantum sensing and efficient dynamic nuclear polarization.

In Chapter 3, A series of metal-organic frameworks (MOFs) assembled with diazatetracene (DAT)-based linkers are synthesized and characterized. Despite different chromophore orientations and spacings, photoinduced persistent radicals were generated in all the MOFs, and their spin-lattice relaxation time (T_1) and spin-spin relaxation time (T_2) are found to be relatively long even at room temperature. The generality of long T_1 and T_2 of photogenerated radicals in the chromophore-assembled MOFs provides a new platform towards quantum sensing applications.

The achievement of extremely long spin polarization in MOFs with nanopores accessible to guest molecules will be an important cornerstone for future highly sensitive quantum sensing and efficient dynamic nuclear polarization.