Ab-initio studies of molecular magnets for quantum information science applications

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Magnetic molecules have been proposed to be versatile for potential magnetic storage, molecular spintronics, quantum sensing, quantum register, and quantum computing applications, by modifying the local chemical environments. Compared to solid-state systems where many practical advances have been made for these applications, magnetic molecular systems are currently being explored to identify ideal molecular candidates and corresponding robust chemical environments for such applications. One popular approach is to design molecular systems whose properties are analogous to the representative solid-state systems for the applications. A few examples are Cr-based molecules in which electron spin qubits were optically addressed and controlled, and rare-earth-based magnetic molecules in which entanglement of nuclear spin qubits was achieved via electric or optical means. In all these systems, electron or nuclear spin qubit states are provided by 3d or 4f electrons which are strongly correlated and/or can form multiconfigurational states. Therefore, an understanding of their properties in the ground and excited states requires ab-initio methods beyond density-functional theory. In this talk, we present our calculated ground- and excited-state electronic and magnetic properties of several molecular systems which passed proof of concept tests for quantum computing applications, such as Cr-based molecules, rare-earth-based molecules, and magnetic adatoms on an insulating substrate, by using ab-initio multireference methods including spin-orbit coupling and relativistic hyperfine coupling. Our results will be useful for designing magnetic molecular systems for quantum information science applications.