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Title:	SPIN- POLARIZED ELECTRON TRANSPORT THROUGH ORGANIC MAGNETIC MOLECULES
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Abstract:	<p>The generation of spin polarized current and its transportation through molecular-species is a pre-requisite for spintronic devices delivering metallic features for one spin channel while insulating for the other. The primary aim of the thesis is to uncover the potential of Organic Magnetic Molecules (OMMs) for spintronic applications. The first principle description of spin polarization across metal-molecule-metal junction in general is not trivial and there are many open questions regarding the effect of presence of localized spin centers on electron transport through a molecular junction. In this thesis, we focus on the fundamental understanding of spin-polarized quantum transport phenomena mediated by organic radicals and proposal of novel strategies to incite sustainable spin polarization in molecular devices. The rational design and synthesis of thermally stable OMMs exhibiting strong magnetic exchange interactions (2J) has always remained a challenge due to transient nature of radical species. At first, we have investigated the electronic and magnetic properties of one of the recently emerged and thermally stable 1,2,4-benzotriazinyl (Blatter's) radical with the simultaneous modelling of several Blatter's based diradicals exhibiting strong ferromagnetic exchange interactions. Using a genre of DFT and wavefunction based CASSCF/NEVPT2 methods, we have introduced X- annulation as a novel strategy to reach even higher values of 2J with the simultaneous rigidification of molecular structure. In the view of device applications, much cultivated stable organic radicals are then incorporated in a single molecule junction setup between two gold electrodes. Employing non-equilibrium Green's function (NEGF-DFT), we investigated the effect of localized spin centers on electron transport through molecular junctions. Quantum Interference (QI) effects play a crucial role in controlling the molecular conductance by allowing or prohibiting the electron to propagate across junction via constructive and destructive QI effects. We demonstrated that incorporating a radical center manipulates the QI features in such a way that meta coupled radicals yield high conductance as compared to para ones. This scenario is in complete contrast to non-magnetic molecules where para coupled molecules are predicted to be better conductors. Apart from the anomalous QI features, the spin filtering efficiency of several radicals attached to the framework of pentacene is investigated. We show that presence of unpaired electron is not a sole requirement to incite spin polarization in junctions. Several factors inherited from the electronic structure of radical moiety plays a decisive role in inducing spin filtering effects. In another approach to achieve ultrafast control over spin polarization, we have proposed a stimuli-based spin filter by utilizing a photo-responsive endoperoxide (EPO) based single molecule device. The photo-irradiation of EPO triggers the homolytic cleavage of the peroxide O-O bond generating a diradical intermediate centred on two O-atoms which facilitates high spin filtering efficiency when placed between gold electrodes. The broken conjugated scenario due to the peroxide bridge of EPO hinders the propagation of de Broglie waves across the molecular skeleton. On the other hand, the diradical intermediate of EPO yields high conductance for one of the spin configurations. The key role played by quantum interference(QI) effects in the dramatic modulation of conductance arising due to different degrees of conjugation along the photochemical reaction pathway of EPO is demonstrated. Finally, with an objective to induce spin polarization for long range electron transport, we employed higher order polyacenes and cumulenes with intrinsic open shell ground state. Apart from the spin filtering effects, we investigated the length dependence of the conductance for aforementioned molecules with pronounced diradical character. The recent accomplishments in the design of molecular nanowires characterized by an increasing conductance with length, in violation to classical Ohm's law, has embarked the origin of "anti-ohmic" wires. This highly desirable behaviour for long-charge transport is investigated within unrestricted DFT formalism with spin and spatial symmetry breaking. The contrasting features of transmission spectra in closed shell (CSS) and open shell (OSS) state eventually evolves in the form of inverted attenuation factors in CSS and OSS ground state. An increase in conductance is observed in CSS state, while the onset of OSS ground state with the increasing molecular length enforces the regular decay of conductance. We postulate the preferential existence of molecule in CSS ground state as a guiding rule to observe the desired anti-ohmic behavior</p>
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