

Insights on Spin Polarization through the Spin Density Source Function

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ABSTRACT : Understanding how spin information is transmitted from paramagnetic to non-magnetic centers is crucial in advanced materials research and calls for novel interpretive tools. Herein, we show that the spin density at a point may be seen as determined by a local Source Function for such density, operating at all other points of space.^[1,2] Integration of the local Source over Bader's quantum atoms measures their contribution in determining the spin polarization at any system's location. Each contribution may be then conveniently decomposed in a *magnetic* term due to the magnetic natural orbital(s) density and in a *reaction* or *relaxation* term due to the remaining orbitals density.^[2] A simple test case, ³B₁ water, is chosen to exemplify whether an atom or group of atoms concur or oppose the paramagnetic center in determining a given local spin polarization. Discriminating *magnetic* from *reaction* or *relaxation* contributions to such behaviour strongly enhances chemical insight, though care need to be paid to the large sensitivity of the latter contributions to the level of the computational approach.^[2] Comparison of Source Function atomic contributions to the spin density with those reconstructing the electron density at a system's position, enlightens how the mechanisms which determine the two densities may in general differ and how diverse may be the role played by each system's atom in determining each of the two densities. These mechanisms reflect the quite diverse portraits of the electron density and electron spin density Laplacians, hence the different local concentration/dilution of the total and (α - β) electron densities throughout the system. Being defined in term of an observable, the Source Function for the spin density is also potentially amenable to experimental determination, as customarily performed for its electron density analogue.^[3]

[2] C. Gatti, A. M. Orlando, L. Lo Presti, *Chemical Science*, under revision

[3] C. Gatti, *Struct. Bond.*, 2012, **147**, 193–286