

Microsymposium

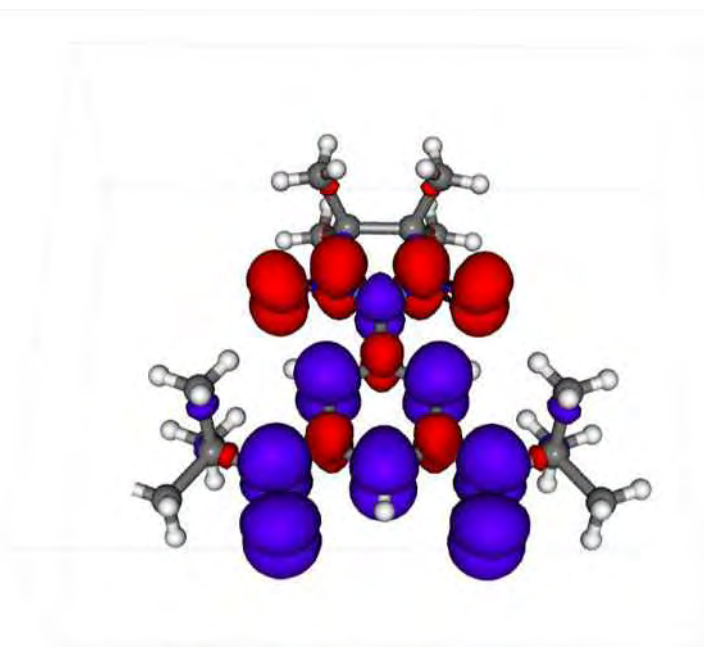
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Purely organic magnets: How to get ferromagnetic interactions

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Purely organic magnets, materials with spontaneous magnetization despite not containing magnetic ions, are very promising for technological applications due to their peculiar properties as flexibility, lightness or even biocompatibility. Most of the published purely organic magnets are free radical compounds. One of the most important difficulties in order to get spontaneous magnetization in such materials is that magnetic interactions among free radicals are usually antiferromagnetic and, in addition, ferromagnetic interactions are weaker than the antiferromagnetic ones. A possible strategy to overcome this issue is the use of triradical molecules with total spin $S=1/2$. In this case, an adequate packing of the triradical molecules can give place to antiferromagnetic interactions between regions with positive spin density and regions with negative spin density of two close molecules. This antiferromagnetic interaction between regions with opposite spin density would result in an overall ferromagnetic interaction between the two close triradicals. With this idea in mind we have performed a study of the spin density distribution and of the intramolecular and intermolecular magnetic interactions of the triradical compound 2-[3',5'-bis(Ntert-butylaminoxyl)phenyl]-4,4,5,5-tetramethyl-4,5-dihydro-1H-imidazol-1-yl-3-oxide, containing two N-tert-butyl aminoxyls and a nitronyl nitroxide groups. Combination of experimental data from a polarized neutron diffraction experiment and ab initio calculations (DFT) has allowed us to obtain the spin density distribution. In addition, the intramolecular and intermolecular magnetic interactions have been computed by ab initio quantum chemistry methods. The values for the intramolecular interactions confirm the $S=1/2$ ground state of the triradical. As for the intermolecular interactions, the two strongest ones are ferromagnetic, what is in agreement with the overlapping of regions with opposite spin density of the two interacting triradicals. These results support the strategy of using triradical molecules for obtaining purely organic magnets with higher magnetic transition ordering temperatures since it is easier to obtain ferromagnetic interactions between the radicals and these interactions, having an antiferromagnetic origin, can be stronger than typical ferromagnetic interactions between radicals.



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