MS108.O01

joint-refinement of spin and charge densities of organic radicals:

Mohamed Souhassou[1], Ariste Bolivard Voufack[1], Nicolas Claiser[1], Claude Lecomte[1], Béatrice Gillon[2], Zeyin YAN[3], Jean-Michel Gillet[3], Marco Marazzi[4], Alessandro Genoni[4]

[1]CRM2, Université De Lorraine, Vandoeuvre-les-nancy, France, [2]LLB, UMR 12, CEA Saclay, Gif sur-Yvette cedex, France, [3]CentraleSupelec, Grande Voie des Vignes, Chatenay-Malabry Cedex, France, [4]SRSMC UMR 7565 CNRS, Université de Lorraine, Vandoeuvre-lès-Nancy, France

E-mail: mohamed.souhassou@univ-lorraine.fr

Joint refinement of X-ray and polarized neutron diffraction data has been carried out in order to determine simultaneously charge and spin density distributions[1] on organic free radicals (2-(4-thiomethylphenyl)-4,4,5,5-tetramethylimidazoline-1-oxyl-3-oxide, Nit(SMe)Ph), which belongs to the nitronyl-nitroxide free radical family (ferromagnetic transition at 0.2 K) The unpaired electron is located on nitronyl nitroxide[3] function. This radical is widely used in the design of molecular magnet because it can propagate magnetic interactions from one molecule to another in the crystalline solids when the packing is adequately tuned. Hydrogen bonding can further control the crystalline molecular arrangement, and therefore direct through space magnetic coupling in the crystal, leading in some cases to ferromagnetic ordering. To assess the importance of intermolecular interactions on magnetic coupling in organic ferromagnets, experimental charge and spin densities obtained from joint refinement[2] will be compared to high level DFT and CASSCF calculations. We will show that DFT fails to reproduce some subtle electron and spin density features contrary to CASSCF ones

1. M. Deutsch, B. Gillon, N. Claiser, J.-M.l Gillet, C. Lecomte, and M. Souhassou, IUCrJ. 2014 May 1; 1(Pt 3): 194–199.
2. A-B. Voufackb, N. Claiser, C. Lecomte, S. Pillet, Y. Pontillon, B. Gillon, Z. Yan, J.-M. Gillet, M. Marazzi , A. Genoni, and M. Souhassou. Submitted to Acta. Cryst. B
3. Pontillon, Y., Caneschi, A., Gatteschi, D., Grand, A., Ressouche, E..S.R. & Schweizer, J. (1999). Chem. Eur. J. 5, No 12, 3616—3624.

Keywords: charge and spin densities, X-ray and polarised neutron , organic radicals