**COUPLED CLUSTER EVALUATION OF THE FREQUENCY DISPERSION OF THE FIRST AND SECOND HYPERPOLARIZABILITIES OF WATER, METHANOL AND DIMETHYL ETHER**

Pierre Beaujean and Benoît Champagne

*Laboratory of Theoretical Chemistry — University of Namur
61, Rue de Bruxelles, B-5000 Namur
pierre.beaujean@unamur.be*

 The static and dynamic first (β||) and the second (γ||) hyperpolarizabilities of water, methanol, and dimethyl ether have been evaluated within the response function approach [1,2] using a hierarchy of coupled cluster levels of approximation and doubly-augmented correlation consistent atomic basis sets. For the three compounds, the electronic β|| and γ|| values calculated at the CCSD and CC3 levels are in good agreement with gas phase electric field-induced second harmonic generation (EFISHG) measurements [3,4]. In addition, for dimethyl ether, the frequency dispersion of both properties follows closely recent experimental values [5] demonstrating the reliability of these methods and levels of approximation. This also suggests that the vibrational contributions to the EFISHG responses of these molecules are small.

[1] C. Hättig and P. Jørgensen, Theo. Chem. Acc. 100 (1998), 230
[2] C. Hättig and P. Jørgensen, Adv. Quant. Chem. 35 (1999), 111
[3] J.F. Ward and C. K. Miller, Phy. Rev. A 19 (1979), 826
[4] P. Kaatz, E. Donley, and D.P. Shelton, J. Chem. Phys. 108 (1998), 849
[5] V. W. Couling and D. P. Shelton, J. Chem. Phys. 143 (2015), 224307